ISOTOPES AND RADIATION TECHNOLOGY

A Quarterly Technical Progress Review

Prepared for DIVISION OF TECHNICAL INFORMATION / DIVISION OF ISOTOPES DEVELOPMENT, UNITED STATES ATOMIC ENERGY COMMISSION, by P. S. BAKER, A. F. RUPP, and ASSOCIATES, OAK RIDGE NATIONAL LABORATORY

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ISOTOPES AND RADIATION TECHNOLOGY

A REVIEW OF RECENT DEVELOPMENTS

Prepared for DIVISION OF TECHNICAL INFORMATION / DIVISION OF ISOTOPES DEVELOPMENT, UNITED STATES ATOMIC ENERGY COMMISSION, by P. S. BAKER, A. F. RUPP, and ASSOCIATES, OAK RIDGE NATIONAL LABORATORY

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Foreword

Isotopes and Radiation Technology is a Technical Progress Review prepared by the Isotopes Development Center, Oak Ridge National Laboratory, at the request of the Divisions of Technical Information and Isotopes Development, U. S. Atomic Energy Commission. This Review is intended to assist those interested in keeping abreast of significant developments in the fields of isotopes and radiation technology. It is not a comprehensive review of all literature published in this field during a given quarter; rather, it is a mechanism for presenting concise, selected reviews of information on subjects of prevailing Commission interest as it becomes available.

This Review attempts to relate the published results of research and development sponsored by the Division of Isotopes Development to significant developments in radioisotopes and radiation technology, as reported in the world literature. Coverage includes isotope production and development, isotope technology development, isotope applications technology, process radiation development, and radiation processing of food and medical supplies.

In addition to reviews of current literature and special review articles dealing with specific isotopes, facilities, and applications, this publication occasionally contains feature articles prepared by recognized experts on specific topics of current interest. Critical evaluations and interpretations presented are those of the editors and invited reviewers; therefore readers are encouraged to consult the original references in order to obtain all the background of the work reported and the interpretation of the results given by the original authors.

The subject headings listed below have been adopted to help maintain continuity and order, from one issue to the next, in the material reviewed. All reviews, except Feature Articles, will be arranged under these headings; but any one issue will not necessarily contain all the headings.

Isotope Production and Development
Isotope Technology Development
Food Irradiation
Process Radiation Development
Source Development
Isotopic Power Development
Applications in Medicine
Safety
Miscellaneous Items of Interest

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Section

I

Isotopes and Radiation Technology

Isotope Production and Development

Periodic reviews of facilities for producing and using isotopes are an important part of the coverage of *Isotopes and Radiation Technology*. In the past, articles have covered such topics as the Oak Ridge National Laboratory (ORNL) electromagnetic separations facilities,[1] research reactor,[2] and 86-in. cyclotron,[3] the Brookhaven National Laboratory (BNL) High Intensity Radiation Development Laboratory,[4] and the AEC–Bureau of Commercial Fisheries' Marine Products Development Irradiator.[5] Future issues will include discussions of other facilities, including machine and radioisotope irradiators and other production plants. The following article on the Hanford Operations is presented as one of the continuing series of discussions.

Hanford Isotopes Plant

By C. A. Rohrmann

Introduction

Radioisotopes have gained wide recognition for their unique advantages as noninterruptable, highly reliable heat and power sources. The practicality of radioisotopic power has been convincingly demonstrated by successful performance both in terrestrial applications (arctic and antarctic unmanned meteorological telemetry stations) and in space (Transit satellites). Other applications under study include space propulsion motors of low to moderate thrust, environmental temperature control for space systems, undersea telemetry for navigation and monitoring, and advanced scientific satellites. Recent AEC reports have covered these requirements and potential applications.[6]

Existing facilities at Hanford, operated by the General Electric Company for the AEC, are being used to provide relatively large, but not unlimited, quantities of crude and purified fission-product concentrates of ^{90}Sr, ^{137}Cs, ^{144}Ce, and ^{147}Pm. These products have been shipped as liquids, dried powders, or granular solids in massive and costly containers to Oak Ridge, Tenn., or to Quehanna, Pa. There, in large experimental facilities, they are further purified and finally encapsulated to provide a variety of sources, including power sources. Realization of the full potential of these sources would be enhanced by the lower costs attainable through large-scale integrated production of securely encapsulated materials at the source of supply. The present Hanford Waste Management Program provides an opportunity for recovery of fission-product isotopes from resources currently treated as waste.

HANFORD WASTE MANAGEMENT PROGRAM

Chemical processing of both stored and current Hanford fission-product wastes is planned and will make the four radioisotopes of interest in this study available in a partially purified and relatively concentrated form. This practical approach to the confinement of radioactive wastes provides the most efficient utilization of an existing substantial investment in waste storage facilities.[7]

Basically, the program entails separation of wastes into fractions of different heat-generation characteristics. One fraction includes those fission products (^{90}Sr and ^{137}Cs) which generate heat for a long time. This fraction will be stored in concentrated form (as dried sorbates on a synthetic zeolite) in high-integrity containers. The short-lived fission products together with the nonradioactive waste chemicals constitute the other fraction. After a suitable period (a few years) of interim storage, this fraction can be safely evaporated to a solid salt cake and stored in existing underground waste

tanks. The transition from storage as rather dilute solutions to storage as more compact solids substantially increases storage capacity without additional investment in storage tanks. Removal of cesium and strontium is essential to avoid exceeding the heat-dissipation capabilities of a solid-filled tank. The length of the interim solution storage is governed largely by the rare-earth fission products; they represent the predominant heat-generating components.

Removal of rare-earth fission products is planned in order to schedule most efficiently the transition to solid-filled tanks. They will be held separately in interim liquid storage so that solidification of bulk wastes can be started earlier. On a steady-state basis, cooled "early-generation" rare-earth fractions will be blended and solidified with "later-generation" bulk wastes.

The planned processing of wastes for the Hanford Waste Management Program, therefore, will make cesium and strontium (blended but relatively free of other fission products) and the rare-earth fission products (including both cerium and promethium) conveniently available as partially purified and relatively concentrated solutions ideally suited for further processing in the Hanford Isotopes Plant.

AEC Criteria for Processing Plant

In view of the above considerations, the AEC in October 1962 requested[8] that the General Electric Company develop a conceptual design and estimates of capital, operating, and unit costs for a processing plant to be located at Hanford. This plant would be capable of high-volume production of encapsulated heat-source forms of ^{90}Sr, ^{137}Cs, ^{144}Ce, and ^{147}Pm. The conceptual design and estimates of capital and operating costs were presented[9] in a report issued in July 1963 and provide sufficient detail for use as the basis for scope design. This present manuscript is essentially a condensation of that report.

The following stipulations were made by the AEC and served as the bases for this study:

1. *Plant Site.* The plant would be located on the Hanford reservation, on a site optimized from the standpoint of essential tie-in with other Hanford facilities but allowing geographic and administrative separation.

2. *Plant Operation.* The plant would be operated independently of other Hanford facilities with the exception of dependence upon other Hanford facilities and operations for feed, disposal of waste, and the provision of certain utilities.

3. *Feed.* The feed to the plant would comprise materials produced by (and optimized for) the Hanford Waste Management Program.

4. *Product and Production Capability.* The plant would permit simultaneous annual production, in four separate production lines, of encapsulated heat sources from

10 Mc of ^{90}Sr as the titanate ($SrTiO_3$)
10 Mc of ^{137}Cs as a borosilicate glass
100 Mc of ^{144}Ce as the oxide (CeO_2)
30 Mc of ^{147}Pm as the oxide (Pm_2O_3)

and, on demand, for preparation of any or all of the four products as purified bulk products or intermediates.

5. *Waste Disposal.* The plant would produce wastes compatible with the equipment, facilities, and practices of the Hanford Waste Management Program to which such wastes will be delivered for disposal.

6. *Technology.* The plant would use existing technology or modest modifications thereof. Cost factors offering significant promise of reduction through advancements in technology should be identified.

7. *Product Delivery.* Provisions should be made for delivery of products by in-plant loading of encapsulated sources into customers' devices or customers' shipping casks, or by loading of purified bulk products or intermediates into customers' shipping casks.

Within the context of the above guidelines, the conceptual design seeks to achieve appropriate cost benefits resulting from high throughput; multiple-product output through joint use of the same facilities where economically attractive and technically feasible; and production of a minimum realistic variety of standardized heat-source forms and geometries, while preserving sufficient flexibility to accommodate changed specifications in composition and geometry of product heat sources.

The Products

Selection of an optimum radionuclide for a specific heat- or power-generation use entails comparison of available or attainable materials on the bases of (1) half-life, (2) power density

(watts/cm^3), (3) biological and instrumentation shielding requirements, (4) availability, and (5) cost. Through such considerations the specified four radionuclides were selected as those of most immediate promise and utility for recovery from fission-product wastes.

The properties of the radioisotope forms to be produced are summarized in Table I-1.

Table I-1 PROPERTIES OF RADIOISOTOPIC PRODUCT FORMS

	^{90}Sr	^{137}Cs	^{144}Ce	^{147}Pm
Types of decay	Beta	Beta-gamma	Beta-gamma	Beta
Half life, years	28	30	0.78	2.7
Purity,* wt.%	50	35	18	95
Chemical form	$SrTiO_3$	Borosilicate glass	CeO_2	Pm_2O_3
Isotope in compound, wt.%	24	16	15	82
Density of compound,† g/cm^3	4.6	3.2	6.4	6.6
Specific power of pure isotope, watts/g	0.95	0.42	25.6	0.33
Power density of compound, watts/cm^3	1.05	0.22	24.5	1.8

*These isotopic purity values are regarded as attainable but depend on reactor operating conditions, time since discharge, extent of contamination with naturally occurring stable isotopes, and the effectiveness of the purification process. Slightly higher values may also be anticipated.

†Densities may also vary: for cesium glass, the density shown is actual; for $SrTiO_3$, CeO_2, and Pm_2O_3, it is 90% of theoretical.

STRONTIUM-90

Strontium-90 has a half-life of 28 years. Its fission yield (atoms produced per hundred atoms of ^{235}U fissioned) is 5.8%. In addition, highly radioactive ^{89}Sr and the stable isotopes ^{86}Sr and ^{88}Sr are also produced in smaller, but still significant, fission yields. Also some "natural" strontium may be introduced as a contaminant in the reactor fuel and in fuel-processing chemicals. As with all highly radioactive isotopes today, a practical process by which pure ^{90}Sr may be recovered from such mixtures of strontium isotopes does not exist; therefore the ^{90}Sr recovered will be of the concentration existing in the fuel-processing waste stream. Isotopic analysis has indicated that an isotopic purity of about 50% can be obtained routinely. This value has been accepted as the purity basis to be used to describe the quality to be expected. It is conceivable that either slightly higher or slightly lower values may be realized.

Both short-lived (50-day) ^{89}Sr and ^{90}Sr disappear by beta decay to the respective yttrium isotopes, ^{89}Y (stable) and ^{90}Y (64 hr). Yttrium-90 forms stable ^{90}Zr after emission of an extremely energetic beta particle. Although a very small portion of the disintegrations (^{89m}Y to stable ^{89}Y and ^{90}Y to ^{90}Zr) yields gamma emissions of moderate to high energy, the shielding requirements for ^{90}Sr heat sources are largely determined by the bremsstrahlung from the energetic beta emission from ^{90}Y. For young wastes a large fraction of the heat of radioisotopic decay is derived from ^{89}Sr. Aging for about 1 year from reactor discharge is necessary to provide heat sources characteristic of ^{90}Sr (i.e., < 10% of the heat from ^{89}Sr, and hence not exhibiting the rapid decline in heat output characteristic of short-lived ^{89}Sr). Although ^{89}Sr is initially produced at fairly high fission yields (4.8%), its short half-life assures that only very small quantities will be present after this aging period. Since appreciable aging has already been achieved before final processing, the necessary additional aging can take place either in the bulk form in the early stages of purification or in the final capsule without seriously distorting the final product composition.

Pure ^{90}Sr has a calculated specific power of 0.95 watt/g. The strontium titanate compound form is favored because of its extreme inertness, particularly its resistance to dissolution in either freshwater or seawater. The theoretical density of the titanate compound is 5.1 g/cm^3. The power density of such material for strontium of 50% ^{90}Sr content at 90% of theoretical density should be about 1.05 watts/cm^3. There is some sacrifice in power density because of the rather low concentration of strontium in the titanate compound; however, the 50% isotopic purity provides expectations for only moderately

improved power densities for other possible strontium compounds. Strontium is unusual among common metals in that the density of the metal is much lower than that of its compounds. Therefore some of its compounds (SrF_2, SrO, SrS, etc.) have a greater strontium content (g/cm^3) than the pure metal itself; some of these compounds may also be suitable for use as heat sources.

CESIUM-137

Cesium-137 has a half-life of 30 years. Its fission yield is 6.0%, and it comprises about one-third of the cesium isotopes in reactor fuel-processing wastes. The other fission products are stable ^{133}Cs, 2.3-year ^{134}Cs, and 2×10^6-year ^{135}Cs. The ^{134}Cs content is not expected to be present in sufficient amounts (probably <2%) to distort the heat contribution of ^{137}Cs in the mixture of isotopes. For practical purposes ^{135}Cs can be considered stable. The specific power for ^{137}Cs is 0.42 watt/g. The proposed compound form is a cesium borosilicate glass having a density of 3.2 g/cm^3. The power density of the proposed form of the encapsulated cesium "glass" is about 0.22 watt/cm^3. Although ^{137}Cs is a beta emitter, its 2.6-min daughter, ^{137}Ba, is the source of 0.662-Mev gamma activity. A portion of the decay proceeds directly to stable ^{137}Ba with energetic beta emission (which itself can result in some hard bremsstrahlung). Cesium-137 sources require about the same shielding as ^{90}Sr sources. Biologically, ^{137}Cs is less hazardous than ^{90}Sr. Although the fission yield is high, the low isotopic purity, low specific power, low density of the borosilicate glass, and low cesium content of this compound form result in a heat source of rather low power density—about one-fifth that of strontium titanate.

Strontium-90 and ^{137}Cs are comparable in terms of half-life, abundance, and shielding requirements. These fission-product radioisotopes are therefore envisioned as the prime contenders for long-term terrestrial and undersea "workhorse" uses. With adoption of shield-lightening launch-pad techniques, radiation-resistant instrumentation, etc., they may also be considered for long-life use in space.

CERIUM-144

Although the half-life of ^{144}Ce is less than 1 year (285 days), its high fission yield (5.6% from ^{235}U) and high specific power (25.6 watts/g) make it of considerable interest. The extremely energetic gamma activity of its daughter, ^{144}Pr, and its dilution with stable cerium isotopes are disadvantages. Although the energetic gamma activity is significant from a shielding standpoint, its abundance is rather small. Because of the fairly short half-life of ^{144}Ce, its concentration with relation to stable ^{140}Ce and ^{142}Ce will vary significantly depending on the age of the material since formation in the fission process.

Although 33-day ^{141}Ce is initially produced at high fission yield, its relatively short half-life assures that it will be only an unimportant contaminant at the time of use. Initially, the isotopic concentration of ^{144}Ce (exclusive of ^{141}Ce) obtainable from ^{235}U fission would be about 32.5%; however, with the prolonged exposure of fuel and subsequent aging before processing, lower concentrations must be assumed. Furthermore, the yield from plutonium fission is somewhat less. For the realistic situation then, it is unlikely that the isotopic purity of ^{144}Ce will exceed about 18% at time of use. Even at such dilutions, the power density of the oxide (6.4 g/cm^3 at 90% of theoretical density) is about 24.5 watts/cm^3. The biological hazard of ^{144}Ce is comparatively low—about the same as for ^{137}Cs. Because of both its very high specific power and high fission yield, ^{144}Ce is potentially the cheapest heat source on a watt basis. Applications for this isotope will obviously place a premium on a relatively high heat-generation rate. Use of this heat output for spacecraft propulsion or undersea thermoelectric power devices is a possibility. A special consideration is that some uses will require assembly and application of massive curie amounts over a relatively brief time.

PROMETHIUM-147

Promethium does not occur in nature. In the fission process ^{147}Pm is the only isotope of promethium produced in appreciable yield. Its yield (2.6%) is reasonably high, about half that of either ^{90}Sr or ^{137}Cs, both of which are considered as plentiful fission products. It is therefore regarded as being a fairly available material. Its higher atomic weight reduces the apparent disadvantage of fission yield in comparison with ^{90}Sr. Since it has only one step in its decay scheme to "stable" (10^{11} years) ^{147}Sm, ^{147}Pm is a pure beta emitter. The energy of the beta emission is relatively low; therefore the

bremsstrahlung will have low energy—easily shielded with a minimum of mass.[11] However, in the reactor, energetic gamma-emitting 42-day ^{148}Pm is formed by neutron capture in ^{147}Pm. Although this contaminating isotope is present in small concentrations, it is sufficient to mask the advantage of the pure beta activity of ^{147}Pm. When recovered from fuel of only moderate age (less than a year), promethium has enough gamma energy from ^{148}Pm to require shielding essentially the same as for ^{90}Sr and ^{137}Cs. On such a basis, moderately aged promethium shows no significant advantage over those more prevalent and longer lived isotopes. Some gamma radiation can also be expected from the trace amounts of ^{146}Pm which are present. Promethium-146 has a half-life of about 1.9 years which approximates[12] that of ^{147}Pm. Although precise data on ^{146}Pm are scanty, it has been concluded that the amount present is probably not sufficient to add seriously to the shielding problem with well-aged promethium.

For ^{148}Pm with its 42-day half-life, aging for about one ^{147}Pm half-life is sufficient to reduce the total shielding requirements to about that which would be expected for shielding the bremsstrahlung—a fraction of an inch of lead. This is regarded as a minor shielding requirement and is expected to be met to a large extent by the mass of the usual encapsulating material. This aging step would, however, further reduce the apparent yield of ^{147}Pm. Also, achievement of the highest power densities would require separation of the daughter products accumulated during the aging step.

Promethium has a shorter half-life (2.6 years) than nearly all of the other attractive heat-source isotopes, but it is considered long enough for a number of useful missions. The specific power of ^{147}Pm is 0.33 watt/g.[13,14] The power density of the oxide is expected to be about 1.8 watts/cm^3 at 90% of theoretical density.

In addition, promethium has other characteristics that may be utilized advantageously. For example, as a metal it has a melting point of 1080°C. This is far above that of ^{238}Pu (~640°C) and should therefore result in higher thermal efficiencies without the corrosion problems associated with metals of lower melting point.

Biologically, ^{147}Pm is far less hazardous than any of the other heat-source isotopes that have been considered.

If power density is an important consideration, ^{147}Pm is a superior choice. In the oxide form it has about twice the power density of ^{90}Sr in the titanate form, and eight times the power density of ^{137}Cs in the form of the borosilicate glass. In view of the void-space allowance for helium accumulation with alpha emitters, it is conceivable that the capsule power density of promethium will be equivalent if not superior to ^{238}Pu, particularly if the latter is in the form of the oxide. If its low biological hazard can be taken into account (so that a less chemically stable form, like the metal, can be used), advantages in thermal conductivity and heat-source fabrication may also be realized. Because of its weak external gamma field, ^{147}Pm is a most logical candidate for early use as an auxiliary power source, or simply as a heating source, in space applications.

The cost of ^{147}Pm is higher than either ^{90}Sr or ^{137}Cs; however, its cost is not so high as to impair considerations for use. Chromatographic ion-exchange technology already has been successfully demonstrated at Hanford and Oak Ridge[13,15,16] for the recovery of several thousand curies of extremely pure promethium. The acknowledged high cost and limited availability of heat sources that require minor shielding, such as ^{238}Pu, justify consideration of aged ^{147}Pm, especially since large amounts of ^{147}Pm are already at hand in the form of well-aged processing wastes.

Proposed Plant

FACILITIES DESCRIPTION

Site. The plant (Fig. I-1) is located about 120 yd west of the Hanford Waste Management Facility (221-B Plant) and receives feed from, and returns waste to, this facility through encased underground pipelines. Aside from these lines and certain utility inter-ties, the Hanford Isotopes Plant is geographically and administratively separated from other Hanford facilities and operations. A new railroad spur, about 1800 ft long, and access roads and parking for 10 cars are provided.

Building Description. The conceptual Hanford Isotopes Plant consists of a circular process building 140 ft across and 69 ft in height above grade and an attached single-story building measuring 100 by 120 ft (Figs. I-1 through I-4). The process building contains a circular concrete-shielded structure 103 ft in diameter.

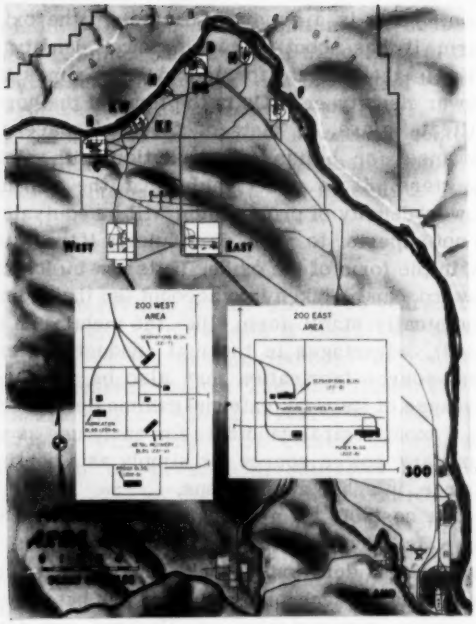

Fig. I-1 Plot plan of the Hanford area.

This structure is divided into 11 manipulator cells and eight process vaults. All of the vaults are equipped with removable cover blocks for top entry by a bridge crane pivoted at the center line of the circular structure.

The plant has a ground floor area of approximately 27,000 sq ft (not including the railroad tunnel), a total floor area of 38,500 sq ft, and a total volume of 1.165 million cubic feet. The process area, which includes the shielded cells, is housed in a domed reinforced-concrete structure with a cylindrical shape and a diameter of about 103 ft. An 18-ft-wide operating gallery, with steel frame and insulated steel siding, surrounds the process area structure. Leading out of the north side is a shielded railroad tunnel. Exhaust ventilation facilities are located south of the building. An office and service area of steel-frame construction with insulated steel siding is located on the west side of the process area (Figs. I-1 through I-4).

The process area includes wet chemistry cells, manipulator cells, and a cell access area surrounded by a service gallery and an operating gallery.

1. *Wet Chemistry Cells.* Eight wet chemistry cells are located below grade (Figs. I-4 through I-6). Six cells are approximately 15 ft deep, and two cells, including the pool cell and the solvent-extraction-column cell, are approximately 26 ft deep. Each cell is accessible from the top by crane through removable concrete cover blocks 5 ft 4 in. thick. Personnel access to all cells except the pool cell is through doors 2 ft 6 in. wide by 7 ft high at the minus 20-ft elevation. At that elevation personnel are shielded from radiation by ordinary concrete walls 5 ft 4 in. thick and cast steel doors about 20 in. thick. The cell floors are lined with stainless steel; the walls and cover blocks are coated with radiation-resistant paint. A chute is provided for transfer of materials from the can decontamination and inspection cell to the pool cell.

2. *Manipulator Cells.* At the main floor level (elevation 0 ft 0 in.) are 13 manipulator cells arranged on the periphery of the cell access area (see Figs. I-3, I-4, I-7, and I-8). Each cell has a clear height of 16 ft. Access is by crane from the top and through doors 2 ft 6 in. wide by 7 ft high in the back of the cell. Material is transferred from the cell access area into the cells through a shielded pass-through located in the back wall of each cell.

Radiation shielding walls on the back side are constructed of ordinary concrete (150 lb/cu ft) 5 ft 4 in. thick. The shielding doors are 20-in.-thick cast steel. Walls on the operating gallery side of the cells are 4-ft-thick high-density concrete (230 lb/cu ft). Dividing walls between cells are 4-ft-thick ordinary concrete. The interior of each cell is lined with stainless steel.

Each cell has one or two 18- by 30-in. viewing windows, depending on the width of the cell. The viewing windows are cold-side loaded, oil-filled lead-glass windows with a shielding density equal to the wall in which they are installed. Three 4-in.-diameter stepped sleeves with shielding plugs are installed through the cell wall in a vertical pattern at each side of each window. Heavy-duty, extended-reach, master-slave manipulators are installed above each viewing window.

A mechanical conveyor is provided for intercell material transfer. Mechanical aspects of the conveyor will be developed in future scope and detail design studies. The conveyor housing within the cells will be shielded with 4 or 5 in. of lead, or equivalent.

(Text continues on page 111.)

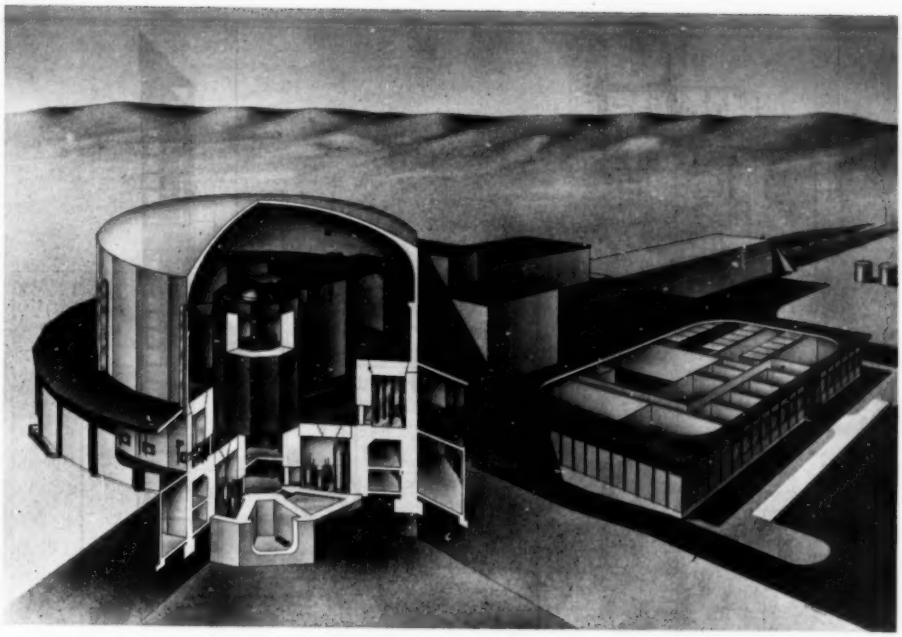

Fig. I-2 Cutaway view of conceptual Hanford Isotopes Plant.

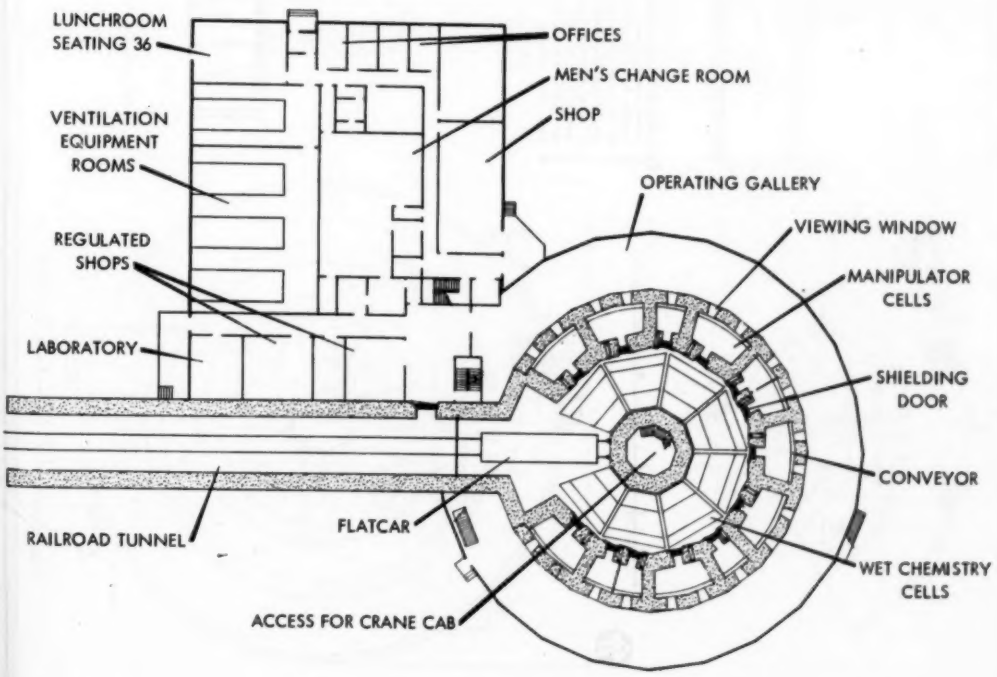

Fig. I-3 Hanford Isotopes Plant floor plan.

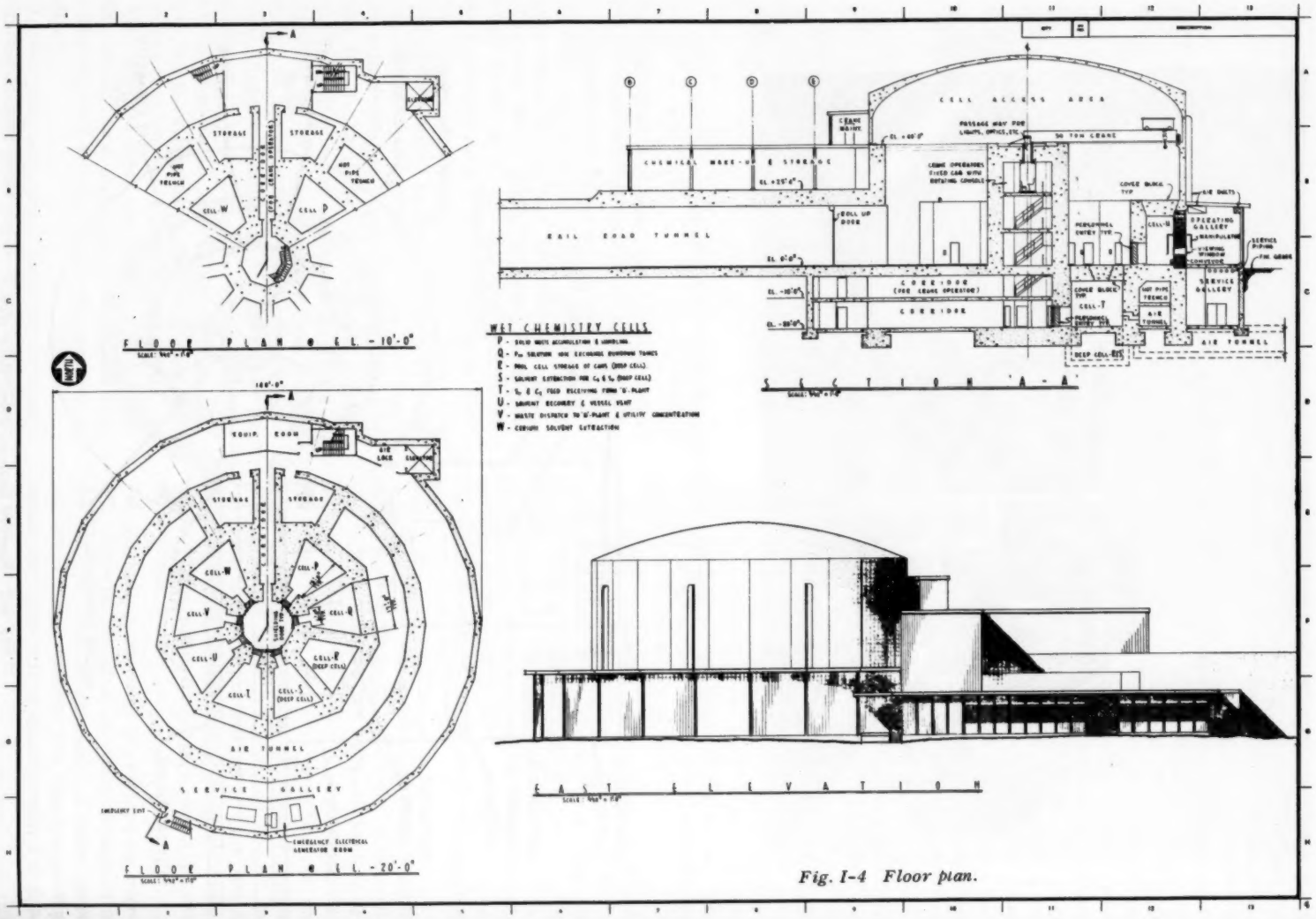
FLOOR PLAN @ EL. -10'-0"
SECTION A-A
WET CHEMISTRY CELLS
P - SOLID WASTE ACCUMULATION & HANDLING
Q - Pu SOLUTION FROM EXCHANGE DOWNFLOW TOWER
E - POOL CELL STORAGE OF CANS (DEEP CELL)
S - SOLVENT EXTRACTION FOR C4 & Pu (DEEP CELL)
T - Pu & C4 FEED RECEIVING FROM 'B' PLANT
U - SOLVENT RECOVERY & VESSEL VENT
V - WASTE DISPATCH TO 'B' PLANT & UTILITY CONCENTRATION
W - CERIUM SOLVENT EXTRACTION
EAST ELEVATION
FLOOR PLAN @ EL. -20'-0"

Fig. I-4 Floor plan.

Fig. I-4 Floor plan.

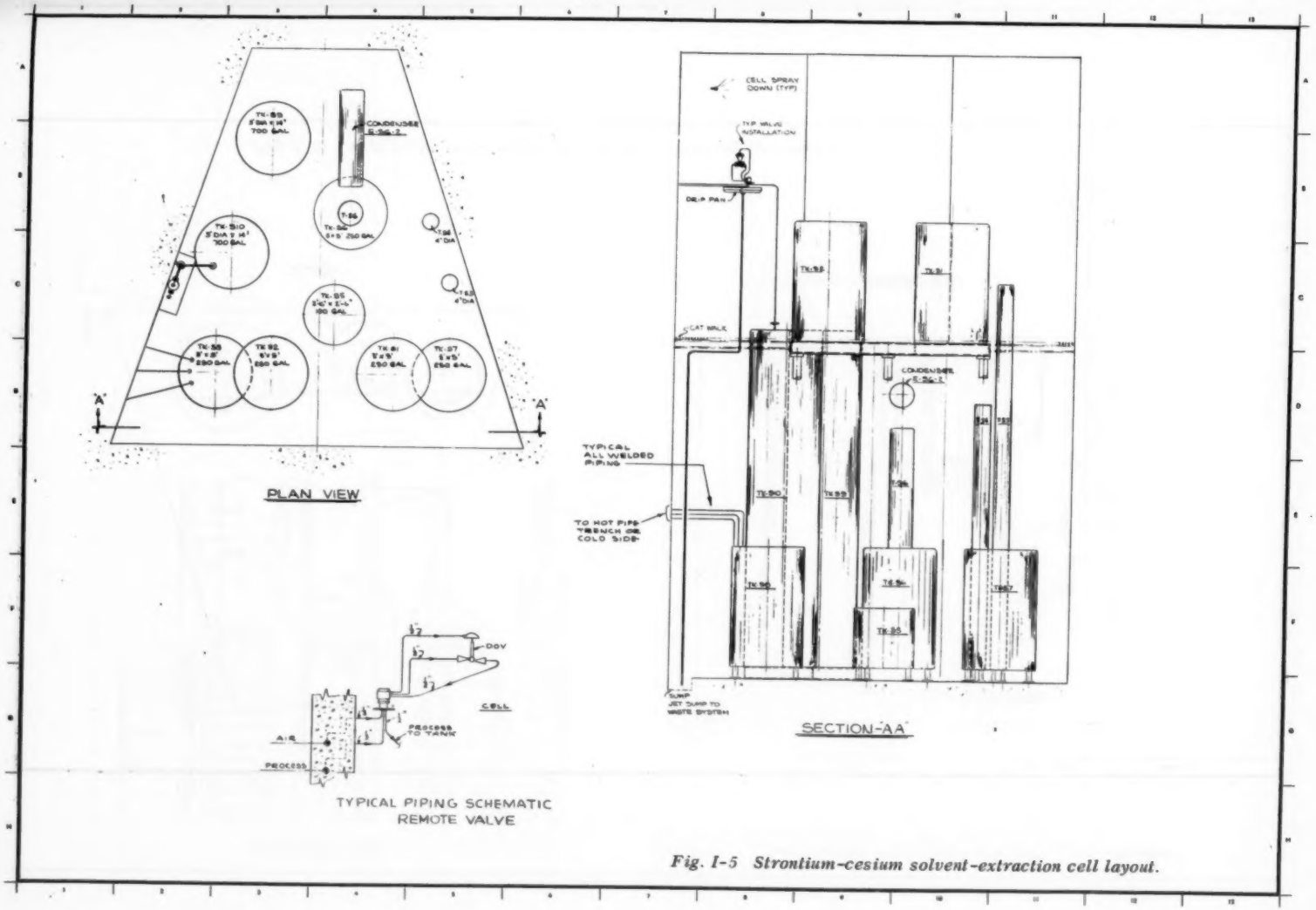

Fig. I-5 Strontium-cesium solvent-extraction cell layout.

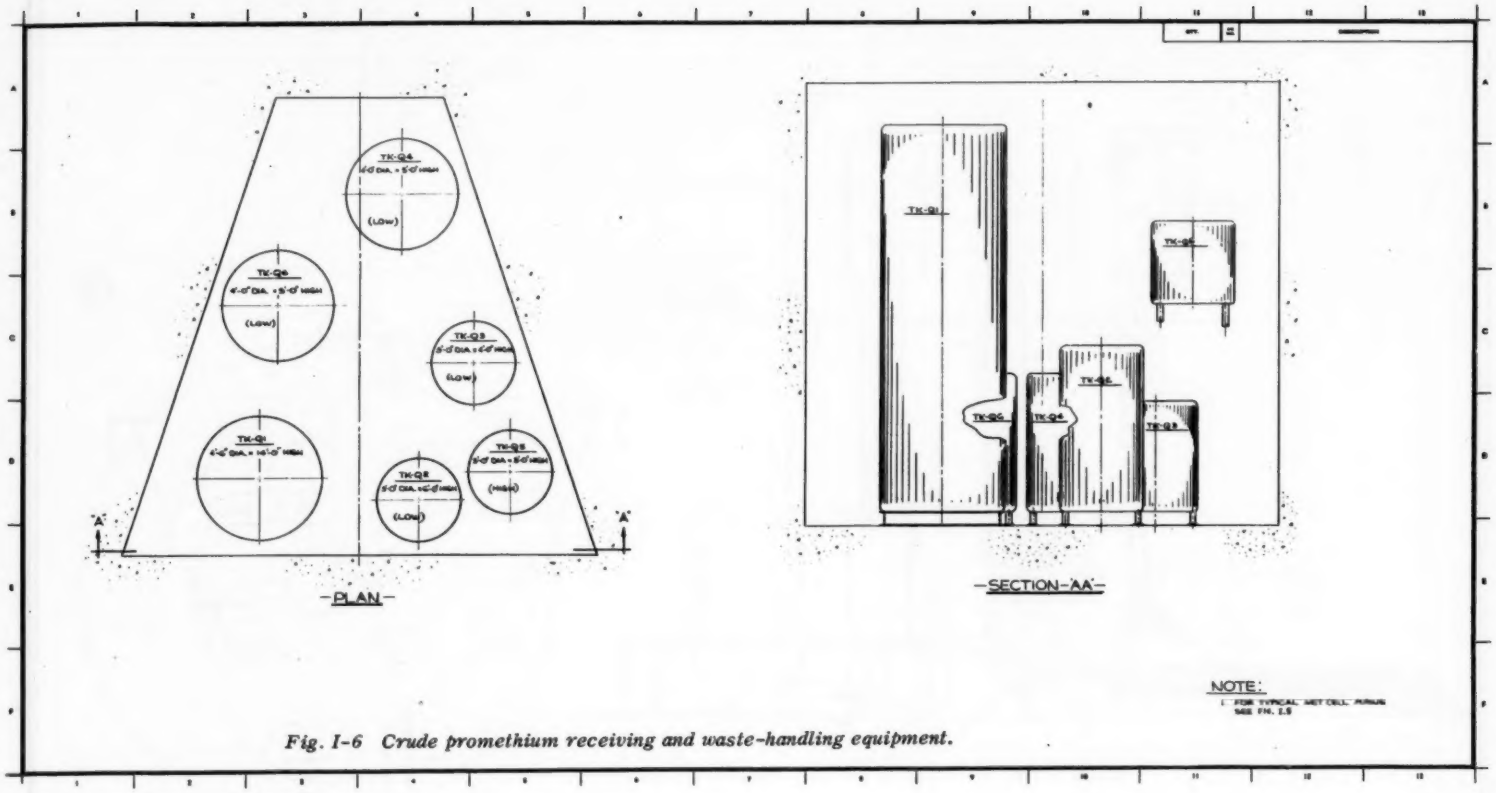

Fig. I-6 Crude promethium receiving and waste-handling equipment.

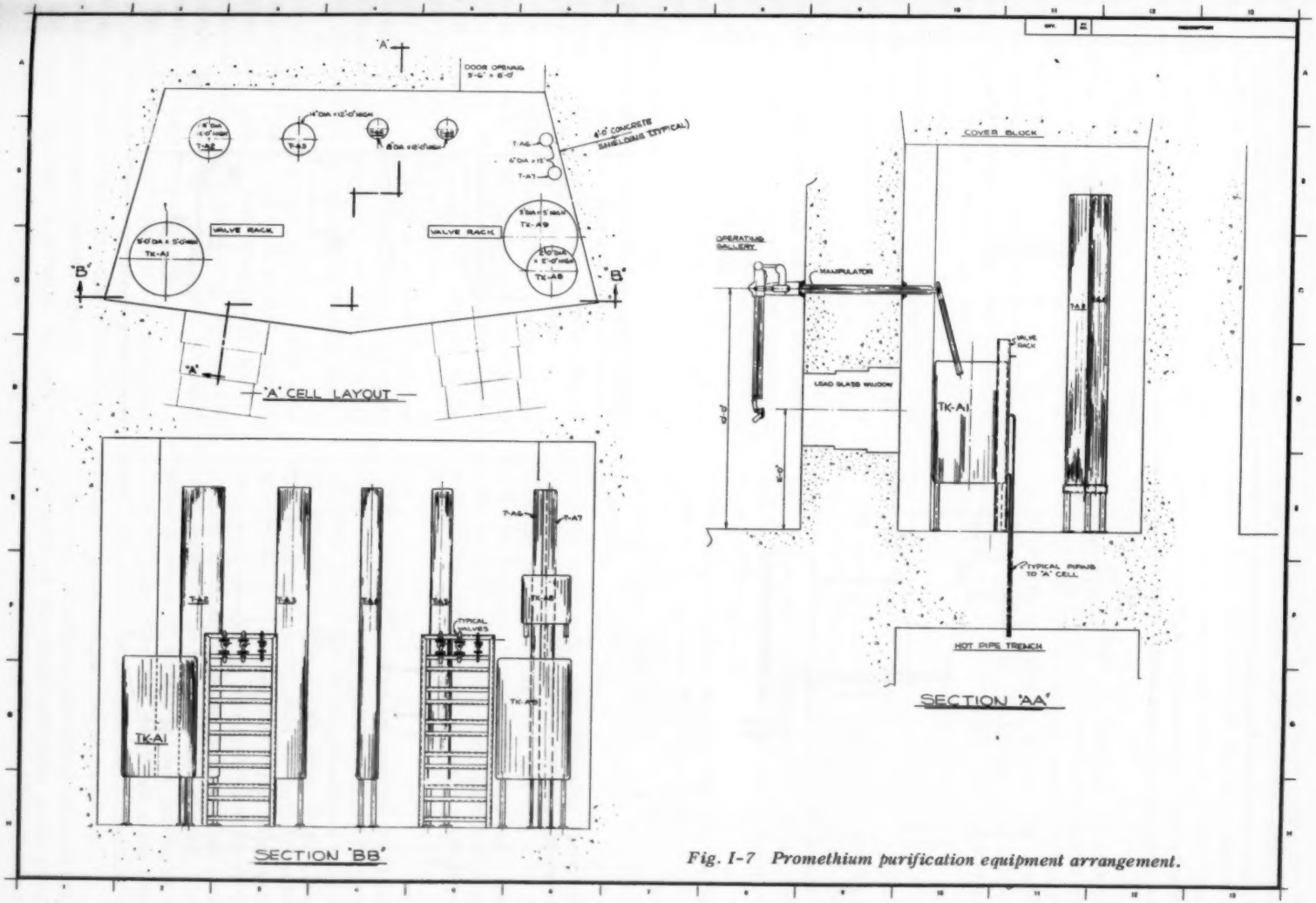

Fig. I-7 Promethium purification equipment arrangement.

Fig. I-8 Equipment arrangement in product packaging cell.

Cell Access Area. A shielded railroad tunnel enters the cell access area for material transfers from the building. Access to the cells is as previously described.

A 30-ton crane, with one 5-ton and two 2-ton auxiliary hoists, is provided for installation and removal of cell equipment. The crane has a double-beam bridge supported on a circular rail at the outer end; the other end is pivoted on a central concrete tower. The control cab is located in a central tower within the concrete shielding walls.

Crane movements of hoist bridge and trolley are performed through stepless variable speeds over a 20 : 1 range. Motors are direct-current type with silicon rectifier power source.

The operator will view the crane operations by periscope optics attached to the crane. The periscope eyepiece has three power ranges and a radial, horizontal, and rotary vertical movement. The operating console is supported from the central bridge turntable and will thus rotate with the crane bridge to give the operator a better "feel" of the work. A separate corridor and access stairwell serve the crane "cab" to permit ready access and to provide fresh air supply.

Lighting fixtures are mounted on the crane in a position to be serviced from the crane bridge. The light is directed downward to illuminate the deep cells and to compensate for the shadow of the crane from other overhead lights.

Communications include one station on the multiple-channel intercommunication and paging system, a two-way intercommunication system to the crane bridge, and one station on the sound-powered telephone system.

All surfaces in the cell access area are coated with a paint that will withstand periodic decontamination. All covers will be overlaid with sheet vinyl to facilitate decontamination.

Service and Operating Galleries. The concrete walls, floors, and ceilings of the service gallery are finished with a paint suitable for decontamination. Partitions are of plastered masonry or concrete, finished with the same materials as the service gallery.

The floor of the operating gallery is covered with sheet vinyl. Concrete, steel walls, and ceiling surfaces in the operating gallery are coated with a chemically resistant paint.

The rooms that have a potential for radioactive contamination are on the process area side of the changeroom. These rooms are the manipulator repair shop, regulated shop, laboratory, storage room, and the crane maintenance room. The concrete floors in this area are covered with sheet vinyl or coated with a paint that can be decontaminated. Partitions are either plastered masonry or metal and are similarly painted.

The remainder of the building includes the changeroom, maintenance shop, storage room, elevator, chemical makeup area, ventilation equipment room, offices, ladies' rest room, and the lunchroom. Partitions are plaster, plastered masonry, or metal, with conventional finishes.

Production Processes

The processes to be employed in the Hanford Waste Management Program and in the Hanford Isotopes Plant have been discussed in detail in earlier publications.[17,18] These processes and their interactions are indicated in Figs. I-9 through I-15.

The Hanford Waste Management process will produce, by solvent extraction (Figs. I-9 and I-10) and ion exchange (in the case of aged alkaline waste supernatants), two streams of interest:

1. An aqueous stream containing a concentrated and partially purified mixture of strontium and cesium at about 98% recovery
2. An aqueous stream containing the rare-earth isotopes including cerium and promethium, with about 90% recovery of cerium and about 80% recovery of promethium

The strontium-cesium concentrate (1BP) will be processed in the Hanford Isotopes Plant by an initial precipitation step to achieve most of the cesium-strontium separation (Fig. I-11). The strontium fraction will be further treated (Fig. I-12) by solvent extraction with di-2-ethylhexyl phosphoric acid (D2EHPA) for purification from other fission products and from nonradioactive inerts such as calcium. This latter process is similar to that developed at ORNL[19–23] and currently employed in the Hanford Strontium Semiworks for production of massive quantities of purified strontium.

Further, strontium processing involves precipitation of strontium carbonate in the presence of a titania slurry, followed by calcination

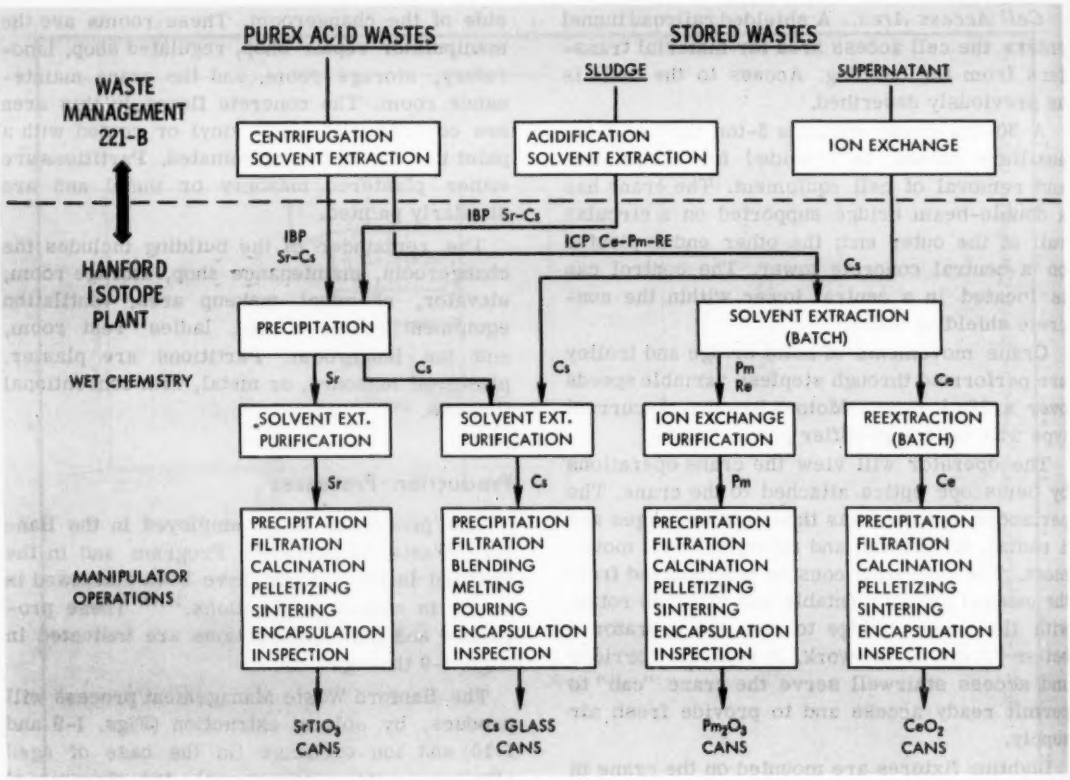

Fig. I-9 Hanford Isotopes Plant flow diagram.

to the titanate, and fabrication into the final heat source.[24–28]

The cesium-bearing supernatant from the precipitation step is processed (Fig. I-13) by solvent extraction for further purification from other fission products and from sodium. The process employs 4-*sec*-butyl-2-(α-methylbenzyl) phenol (BAMBP), a solvent highly selective for cesium. Further cesium processing includes precipitation of cesium oxalate at reduced temperatures, followed by blending with the glass-forming agents boric acid and silica. Subsequent firing at 1100°C results in thermal decomposition of the oxalate and formation of the cesium borosilicate glass.

The cerium–rare-earth fraction from the Waste Management Facility is processed (Fig. I-14) by batch solvent extraction (employing D2EHPA solvent) for isolating cerium (oxidized to the 4^+ state with silver-catalyzed persulfate) from the rare earths. After being stripped from the organic phase, cerium is precipitated as the oxalate for subsequent calcination to the oxide.

The rare-earth fraction containing promethium is further processed by chromatographic ion exchange (Fig. I-15) for separation from other rare earths; it is subsequently precipitated as the oxalate for calcination to the oxide.

Conventional ceramic technology is specified for production of the three proposed refractory products: strontium titanate, cerium oxide, and promethium oxide. Pour casting is assumed for forming of the cesium borosilicate glass. Provision is made for plant loading of encapsulated sources, into either customers' using devices or casks, and for delivery of purified bulk products to customers' casks. Cask and equipment entry is made through a railroad tunnel penetrating the cell structure.

The schedule indicates a beneficial use date of late 1967, which is compatible with that expected for the Hanford Waste Management Facility. This schedule assumes timely authori-

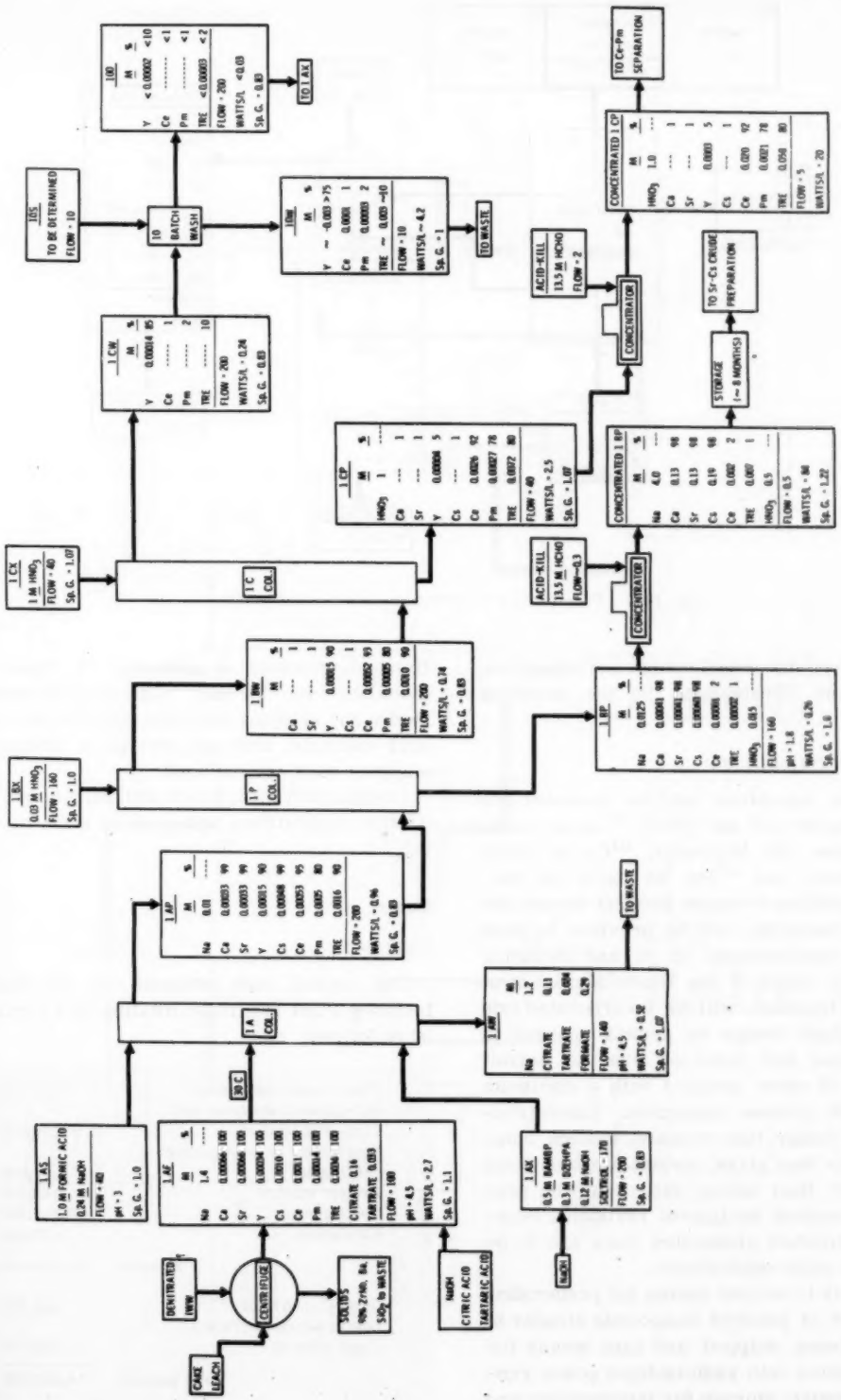

Fig. I-10 Purex denitrated waste processing via CSREX process (an extraction process using D2EHPA–phenol plus a diluent).

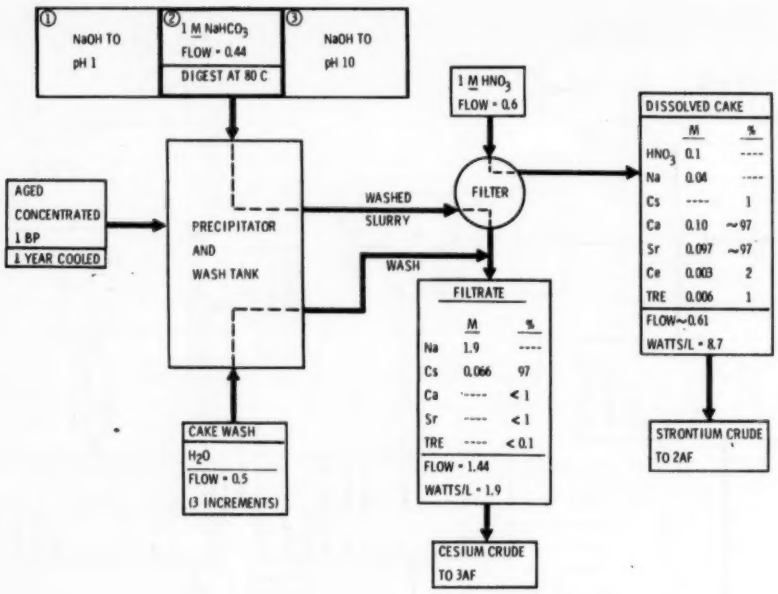

Fig. I-11 Preparation of strontium and cesium crudes.

zation of scope and detail design and supporting research and development by the operating contractor.

PRODUCTION CAPABILITY

Production capability will be provided for ^{90}Sr as titanate (10 Mc/year), ^{137}Cs as borosilicate glass (10 Mc/year), ^{144}Ce as oxide (100 Mc/year), and ^{147}Pm as oxide (30 Mc/year). In addition to these product forms and quantities, flexibility will be provided to meet production requirements up to and including the expected output of the Waste Management Program. Flexibility will be incorporated into the basic plant design to permit changes in product forms and sizes or even to permit processing of other isotopes with a minimum changeout of process equipment. Thus strontium oxide rather than titanate, cesium compounds rather than glass, cerium or promethium metal rather than oxide, etc., could be prepared with modest equipment revisions. Separate and individual production lines are to be provided for each radioisotope.

The plant is to include means for preparation and shipment of purified compounds similar to those now being shipped and also means for loading capsules into radioisotopic power generators. Adequate storage for intermediate and finished products is assumed (2 years' accumulation for ^{90}Sr and ^{137}Cs). Capsule shipping casks and shipping cars are not provided in the cost estimate, although conceptual designs of a shipping cask are included.

Comparison with recent and near-future production capabilities and costs is made in Table I-2.

Costs

CAPITAL

The capital cost estimate for the Hanford Isotopes Plant facilities, totaling $14.3 million, is as follows:

Fixed-price contractor	$ 8,315,000
Management services and off-site inspection	300,000
Supervision of construction (Title III)	250,000
Project startup	165,000
Contingency	2,900,000
Escalation	830,000
Subtotal	$12,760,000
General overhead	340,000
Design services (Title I and Title II)	1,200,000
Total project	$14,300,000

Fig. I-12 Strontium purification and packaging.

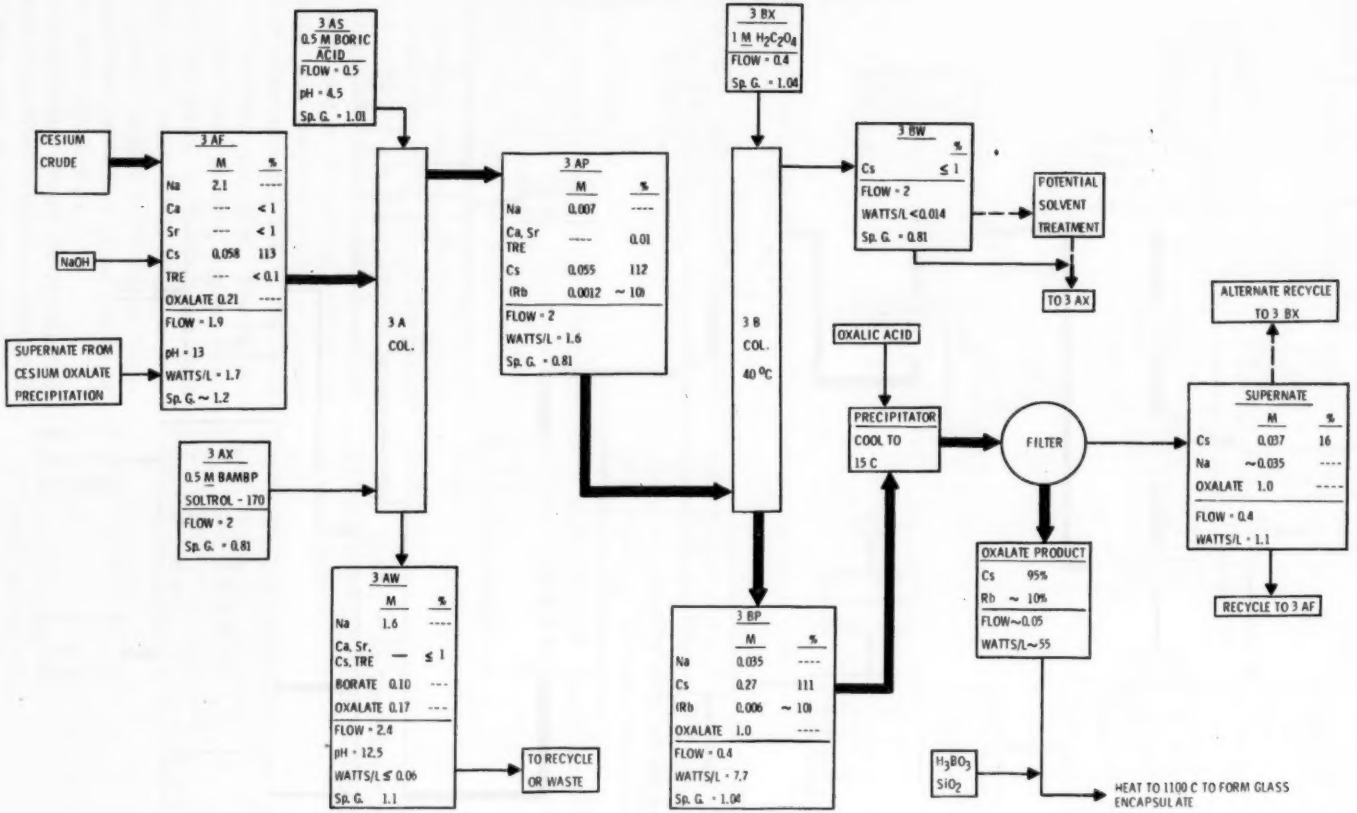

Fig. I-13 Cesium purification and packaging.

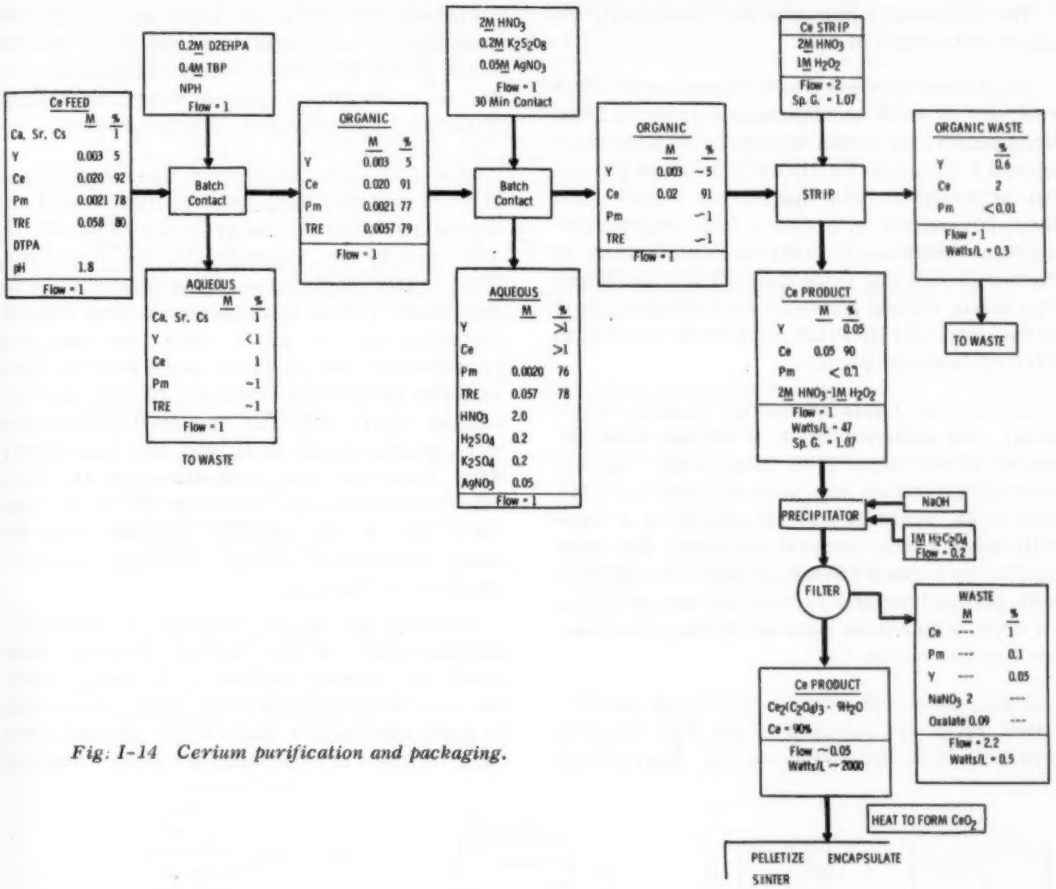

Fig. I-14 Cerium purification and packaging.

A breakdown of the estimated fixed-price contractor's bid price is:

	Fixed-price contractor bid price
Yardwork	$ 180,000
Building and structure	3,000,000
Equipment	3,000,000
Process piping	770,000
Instrumentation	1,200,000
Electrical	165,000
Total	$ 8,315,000

OPERATING

The annual operating cost is estimated to be $4.016 million for operation at the previously indicated design basis of 10 Mc/year of ^{90}Sr as titanate, 10 Mc/year of ^{137}Cs as borosilicate glass, 100 Mc/year of ^{144}Ce as oxide, and 30 Mc/year of ^{147}Pm as oxide; the estimated total staff comprises 64 people on a three-shift 7-day/week basis.

Operating costs included in the schedules in Table I-3 have been estimated on a full operating cost basis, without regard for potential intragovernmental funding implications. It should be noted, however, that (1) no attempt has been made to include a factor to cover current or prior years' research and development or AEC administrative expense and (2) no provision has been included for state and local taxes (other than payroll), indemnification, leasehold, or profit should operations become a commercial enterprise.

These capital and operating cost considerations would result in the unit costs (dollars per curie) to the government shown in Table I-4 (cost breakdown also included).

The following comments describe briefly the major categories of cost:

Feed Material from Waste Management. This category of cost encompasses only those functions performed within the Hanford Waste Management Program Facilities to process chemical separations plant high-level wastes to a form compatible with either final waste-management confinement-program requirements or further processing in the Hanford Isotopes Plant. The dollar amount shown as feed is proportional to the total 221-B Plant production diverted to Hanford Isotopes Plant.

Conversion Costs (Including General Overhead). An assigned staff of 64 has been assumed in development of these costs. The wet chemistry function has been included on a 21-shift/week schedule and all others on a five-shift/week basis. General overhead has been applied on a basis consistent with current Hanford procedures and represents the provision for service functions such as finance, relations, and fire protection.

Depreciation. Standard service-life amortization rates (as outlined in the AEC Manual) have been used in developing the depreciation estimate. The total is made up of $650,000 annually for amortization of the $14.3 million plant plus a $225,000 common-usage allocation for such services as steam, water, electricity, railroad, buses, laundry, and fire station.

Unit Costs for Alternative Operating Plans. It is recognized that, although this report assumes operation at the proposed rates (10, 10, 100, and 30 Mc, respectively, for ^{90}Sr, ^{137}Cs, ^{144}Cs, and ^{147}Pm), other rates may actually be employed. Therefore other cases were studied, including one in which aged (one half-life) promethium was produced along with the other isotopes at the base rates (10, 10, 100, and 15); another where only ^{90}Sr and aged promethium were produced (10, 0, 0, and 15); and another with strontium and promethium at the base rates and cesium and cerium at 20% of the base rates (10, 2, 20, and 30). The unit costs for these alternatives and the base case are summarized in Table I-5.

Although the bases accepted for this engineering study of the Hanford Isotopes Plant would not assure realization of lowest costs, the most significant feature of the results was to show that such a plant would certainly produce isotopes at extremely low costs compared

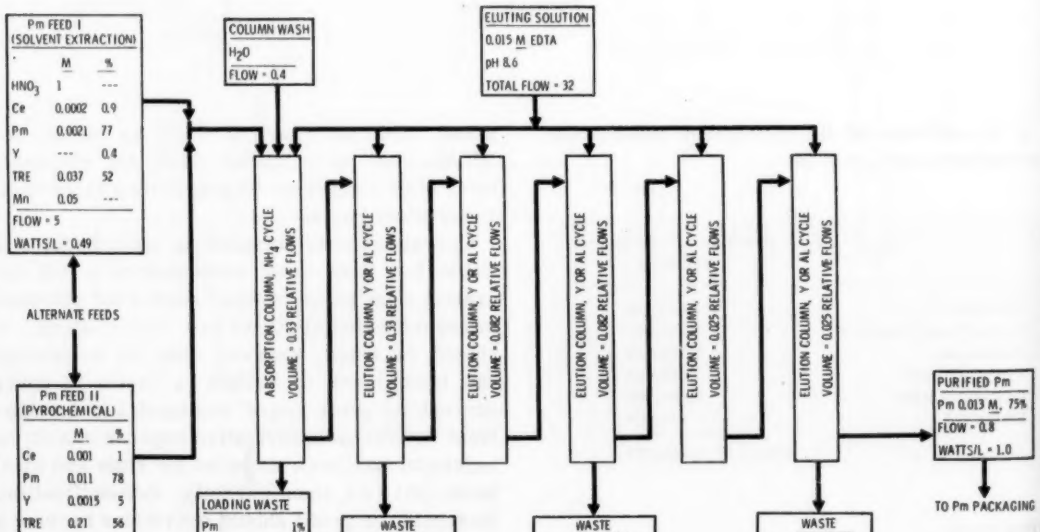

Fig. I-15 Promethium purification and packaging.

Table I-2 PRODUCTION CAPABILITY FOR SELECTED FISSION PRODUCTS

Annual Megacuries and Costs*

	Recent past		Present and near future (interim)		Hanford Isotopes Plant (after FY 1966)	
	Quantity, Mc	Cost/curie	Quantity, Mc	Cost/curie	Quantity, Mc	Cost/curie
^{90}Sr	3	$3.00	5	$1.50	10	$0.127
^{137}Cs	1	3.00	3.5	1.50	10	0.102
^{144}Ce	0.5	3.00	3.5	0.50	100	0.007
^{147}Pm	0	-	0.5	1.00	30	0.033

*Total estimated cost for encapsulated material, using 6-in.-long right cylinders with inside diameters of 1.5, 4, 0.5, and 1.5 in. for ^{90}Sr, ^{137}Cs, ^{144}Ce, and ^{147}Pm, respectively.

Table I-3 HANFORD ISOTOPES PLANT OPERATING COST ESTIMATES

	^{90}Sr	^{137}Cs	^{144}Ce	^{147}Pm	Total
Annual production					
Megacuries	10.0	10.0	100.0	30.0	
Kilowatts (thermal)	66.7	48.3	807	10.8	
Kilograms (pure isotope)	70.4	115.0	31.5	32.8	
Kilograms (product*)	293 ($SrTiO_3$)	719 (boro-silicate glass)	210 (CeO_2)	40 (Pm_2O_3)	
Capsules (sizes as in Table I-2)	363	185	1720	33	
Annual costs (FOB plant sites) (in thousands of dollars)					
Feed materials from Phase III B-Plant	$ 770	$ 550	$ 110	$ 330	$1,760
Hanford Isotopes Plant conversion costs					
Wet chemistry	88	78	85	193	444
Packaging	26	15	133	4	178
Maintenance, engineering, and administration	97	95	108	116	416
General overhead	70	62	108	103	343
Depreciation†	218	219	184	254	875
Total annual costs	$1,269	$1,019	$ 728	$1,000	$4,016
Unit costs					
$/curie	0.127	0.102	0.007	0.033	
$/watt (thermal)	19.03	21.10	0.90	92.60	
$/gram (isotope)	18.03	8.86	23.10	30.60	
$/capsule	3,500.00	5,510.00	423.00	30,300.00	
Kilocuries/capsule	27.6	54.1	58.2	909	
Thermal watts/capsule	184	261	469	327	

*Includes conservative allowance for isotopic purity: ^{90}Sr, 50%; ^{137}Cs, 35%; ^{144}Ce, 18%; ^{147}Pm, 95%.
†Includes $650,000 on facilities (new; estimated on standard AEC Manual service-rate basis) plus $225,000 applicable to existing common-usage facilities.

with today's figures for equivalent materials. It should be emphasized that these costs are for fully encapsulated materials ready for heat-source use. Consideration has also been given to alternative isotope production concepts, along with proposals for processes more closely integrated with the waste-management system, to achieve large reductions in construction costs which are significant cost elements under conditions of private operation for profit.

ALTERNATIVE PLANT DESIGNS

Several options were considered in providing a plant design for the purification and packaging

Table I-4 COST BREAKDOWN FOR ISOTOPE PRODUCTION

Isotope	^{90}Sr	^{137}Cs	^{144}Ce	^{147}Pm
Depreciation (for $14.3 million plant investment)	$0.022	$0.022	$0.002	$0.008
Credit to waste management for feed materials*	0.077	0.055	0.001	0.011
All other manufacturing and overhead	0.028	0.025	0.004	0.014
Total (design basis unit cost)	$0.127	$0.102	$0.007	$0.033

*This cost represents that portion of waste-management costs shared by the Hanford Isotopes Plant when the Waste Management Facility is delivering feed to this plant.

of fission-product isotopes. These alternative paths differ from one another mainly in terms of overall layout of the processing building, plant location, operating philosophy, and, in one case, the number of products. The alternatives are listed below, along with the respective capital cost estimates:

Longitudinal Canyon Type Building. The principal feature of this alternative, as compared to the proposed round building design, is that the building is designed in a rectangular shape and is similar in appearance to the typical "canyon" buildings used in large-scale radiochemical processing. To duplicate the cell floor space, equipment, and overall building volume used in the round building design would require a capital expenditure of $17.0 million, including overhead and design services.

Round Building Alternatives. Round building alternatives included: (1) deferral of installing, initially, equipment required for the packaging of cesium and cerium and (2) construction of a two-product plant—strontium and promethium only.

In the first alternative the installation of cesium and cerium packaging equipment would be made at a later date. Packaging cells would be initially provided as empty cells only. Total initial plant cost for this alternative is $13.6 million.

In the second alternative a round building would be constructed with provision for producing only strontium and promethium. A slight reduction in overall plant size is made, together with elimination of some of the cesium and cerium processing equipment. Total plant cost for this alternative is $11.2 million. Greater cost reductions are not realized by cutting back to two products, because two crude streams are still received from the Waste Management Facility and both must be processed to recover the strontium from one and promethium from the other. Wet chemistry cesium-purification equipment is not totally eliminated since the bulk of this equipment is used for both strontium and cesium purification in the round building plant design. Likewise, in the separation of promethium from cerium, the equipment used for this separation is also used to purify the cerium. So, by eliminating cesium and cerium as final plant products, very little wet chemistry equipment is deleted in this alternative.

Manipulator Annex to B-Plant. An alternative plant design was considered where an addition to the west end of the B-Plant Building would contain manipulator cells having the same amount of manipulator cell space as the round building design. Empty cells in the west end of the B-Plant Canyon would be utilized for the wet chemistry processing required. This alternative would share all utilities, office space, crane, cribs, waste tank farms, and ventilation with B-Plant operations. Capital plant cost for this is $10.4 million.

Manipulator Annex to U-Plant. This alternative plant design is similar to that stated above. However, instead of locating the plant at the west end of B-Plant, it would be located in the west end of U-Plant in the Hanford 200 West Area. Manipulator cells would be added to the end of U-Plant, and the wet chemistry processing operations would be performed in the U-Plant cells. This alternative has the problem of transferring the feed material from its source in East Area to the U-Plant in West Area, a distance of about 5 miles, and also the

Table I-5 HANFORD ISOTOPES PLANT UNIT COSTS FOR ALTERNATIVE CASES

	^{90}Sr	^{137}Cs	^{144}Ce	^{147}Pm
Base case				
Annual production, Mc	10	10	100	30
Unit cost, $/curie	0.127	0.102	0.007	0.033
Aged ^{147}Pm				
Annual production, Mc	10	10	100	15
Unit cost, $/curie	0.127	0.102	0.007	0.065
20% ^{137}Cs and 20% ^{144}Ce				
Annual production, Mc	10	2	20	30
Unit cost, $/curie	0.155	0.142	0.015	0.052
^{90}Sr and aged ^{147}Pm only				
Annual production, Mc	10	0	0	15
Unit cost, $/curie	0.162	-	-	0.083

return of waste streams to the Waste Management Facility in 200 East Area. Capital plant cost for this alternative is $12.0 million.

New, Separate, and Integrated Waste Management and Isotope Production Facility. In view of the close relationship between the Waste Management Program and isotope production, it appeared appropriate to consider the cost of an entirely new, separate, and integrated plant which would combine the function of the two facilities which now form the major basis in the present study. A circular design was chosen, sized, and scaled to provide for the waste processing presently planned for 221-B Plant as well as the isotope production facilities. The estimated construction cost of such a plant is $29.0 million.

It is concluded that this estimate, when compared with the alternative chosen, justifies continued planning along the selected line. This figure also continues to support the great economic advantage in using the existing 221-B structure where possible.

Alternative Products and Processes

As the use of radioisotopic heat and power sources progressively develops, it is expected that the source compound and physical form of these products must change to meet the needs of advanced electric power generator designs and of other applications requiring extremely reliable heat sources. Although the Hanford Isotopes Plant has been designed on the basis of rather specific source compounds and physical forms, the basic facility (purification processes, shielding, access, and equipment maintenance provision, etc.) is inherently adaptable. This is particularly true with respect to the manipulator cells that perform the key function of isotope conversion to final compound and, further, into the final physical form. Thus it is reasonable to assume that alternative source compounds and physical forms could be produced without costly modification.

For example, it is very probable that strontium oxide or fluoride and cesium chloride or sulfate could be prepared and fabricated with only minimal changes in equipment. Furthermore, preparation of metallic, alloy, or cermet forms of strontium, cerium, and promethium could be achieved with modest alterations of final process steps.

Beyond the four fission-product isotopes of immediate concern in this study, the chemical purification technology and mechanical processing means provided in the plant appear adaptable to other radioisotopes. Thus no fundamental aspect of the plant prevents considering large-scale processing of the alpha-emitting heat sources such as ^{228}Th, ^{232}U, ^{242}Cm, and ^{244}Cm. Similar considerations would apply to other fission products such as ^{99}Tc (of no interest as a power source but rather for its cryogenic-superconducting and corrosion-inhibiting properties), and short-lived zirconium, niobium, and ruthenium isotopes.

As isotopic power source and application technology advance, the basic plant design presented here has broad versatility to meet changing needs. Furthermore, the relationship between an isotopes plant and a power-reactor fuel-processing facility is also of special significance. The outputs of the Hanford Isotopes Plant as discussed here could be matched by an annual supply of waste from about 400 tons of 10,000 Mwd per ton of power-reactor fuel (or 200 tons of 20,000 Mwd/ton). Such an output is reasonably certain before 1970. From 400 tons of 10,000 Mwd per ton of fuel at 6 months after discharge, the following contents of other isotopes should be present:

Isotope	Quantity, kg
^{99}Tc	70
^{237}Np	56
^{241}Am–^{243}Am	16 (75+% ^{241}Am and 25–% ^{243}Am)
^{242}Cm	0.82 (convertible to an equivalent amount of ^{238}Pu on aging)
^{244}Cm	0.52

The americium and the curium should appear as discrete bands in the column of the promethium chromatographic ion-exchange system from which they could be recovered by relatively simple means.

Status (October 1964)

Funds have been included in the fiscal year 1965 AEC budget for the "Manipulator Annex to B-Plant" as briefly reviewed under the cost section. The more recent review of this concept indicates a cost of $9.0 million, representing a considerable reduction in construction cost over the case selected for the original study.

In June 1964 the AEC invited the following eight firms to submit bids for the operation of the Hanford chemical processing plants and the Hanford Isotopes Plant: Martin Co., Dow Chemical, U. S. Rubber, National Lead, Allied Chemical, United Nuclear, Monsanto Research, and AMF Atomics. Bids were to be submitted by Oct. 31, 1964, and one of these companies would then be selected for a contract negotiation by about Dec. 31, 1964.

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Section

II

Isotopes and Radiation Technology

Isotope Technology Development

The use of radioisotopes in hydrology is treated extensively in this issue. In past issues two facets of this subject have been reviewed: the use of radioisotopes to study currents at the bottom of the ocean[1] and radioisotope gages to measure snow accumulations.[2] The current discussion includes a recent review of the entire field that was presented to an International Atomic Energy Agency (IAEA) conference in April 1964 and a summary of the IAEA Conference on Hydrology held in Tokyo in March 1963.

This section is concluded with a brief summary of an evaluation of the feasibility of using the Mössbauer effect for solving practical problems in measurement and control systems and a translation of a Russian paper on applications of the Mössbauer effect.

Some Applications of Radioisotope Technology to Water Resources Investigations and Utilization*

By George B. Magin, Jr., and Oscar M. Bizzell†

The U. S. Atomic Energy Commission (AEC) has a broad programmatic interest in radioisotope technology and its application to water-resources investigation and utilization.

At the day-to-day working level, AEC staff members have sought out other agencies and organizations responsible for water-resources development and use and have established cooperative research programs with many of them. The Commission has supplied these agencies (1) knowledge about the capabilities of isotope techniques, (2) special materials needed, and (3) some of the funds.

The United States, with an ideal average rainfall, has vast areas that are arid or semiarid. In other places where water is plentiful, shortages may occur through man's folly or misuse. Improper waste treatment can lead to contamination of both surface and groundwater supplies. Thus problems associated with various U. S. areas range from those of the arid zones where the emphasis is on exploration to those of higher industrialized areas where water demand has overtaken a previously adequate supply. Indeed, hydrologists in this country have been confronted with almost all types of hydrological problems.

The water-resources phase of the Division of Isotopes Development (DID) program was given special impetus by President Eisenhower's address on Aug. 13, 1958, before the United Nations General Assembly meeting concerning Middle East problems. At that meeting he recognized that much new scientific and engineering knowledge of water resources could be obtained through use of isotopes.

Radioisotopes are certainly not a panacea for all the puzzling water-resources problems. However, a number of accomplishments have been made, and the purpose of this report is to describe some of them.

Sediment Density Measurements in Lakes and Streams

The amount of suspended or undissolved solids in streams, rivers, and lakes is of major interest to hydrologists and soil conservationists. The conventional method for these determina-

*This paper was presented at a conference on "Use of Isotope Techniques Applied to Hydrology Problems." A working group met on Apr. 6–10, 1964, in Vienna to coordinate programs in hydrology.

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tions consists of intermittently collecting samples of known volume and separating and weighing the solids. This "grab" sample technique is time-consuming, and measurements are not continuous.

In 1952 the Tennessee Valley Authority (TVA) requested AEC to help develop a radioisotope method for rapid, *in situ* measurements of silt density at the bottom of lakes—at depths to 175 ft. After considerable experimentation with gamma-emitting radioisotopes, Hitch and Bizzell discovered that low-energy bremsstrahlung radiation derived from beta-emitting radioisotopes permitted measurements of silt density with reasonably good sensitivity. This early model was a transmission gage, the first application of bremsstrahlung radiation, and resulted in the award of a U. S. patent.[3]

Parametrics, Inc., Waltham, Mass., has developed an improved sediment concentration gage operating on the same principle. The U. S. Department of Agriculture, U. S. Geological Survey, and other members of the Interagency Group on Sedimentation provided the impetus for this work. The objective was to design and construct a device capable of monitoring undissolved material in the concentration range of 500 to 50,000 ppm. Controlled experiments in simulated streams, performed at the Sedimentation Laboratory at Oxford, Miss., have proved that it meets these specifications.

Initial research indicated that an attenuation gage utilizing the 0.022-Mev X rays from about 1 mc of ^{109}Cd could provide the necessary sensitivity and accuracy. Measuring very small density changes required the use of a reference-cell technique to eliminate errors from changes in natural radiation background, water temperature, and electronic drifts in the measuring equipment. It was noted that a solids concentration change of 500 ppm of typical sediment material represents a change in density of only about two units in the fourth decimal place. This is equivalent to detecting 0.2 mg in a 1-g sample of water, or 0.02%.

An experimental gage (Fig. II-1) was designed. Measurements from either cell were obtained by shifting the position of the ^{109}Cd source. The reference cell, containing distilled water, is isolated from the surroundings, and tests showed that it is an excellent means of compensation for electronic drifts and environmental variations. The test cell, which is opposite the reference cell, is open on three sides to allow easy flow of the water under test. The mean path length of the transmitted radiation in the liquid absorber of either cell is 8.0 cm, and the source is viewed by a 2-in.-diameter NaI scintillation detector at a 55° angle. The concentration of suspended solids is determined by first taking a series of alternate readings of detected radiation in the two cells. Once the ratio of the total test-cell count to the total reference-cell count is established, the relative concentration of suspended solids can be obtained.

A geochemical survey of conditions existing at over 100 sampling stations indicated that normal variations in silt compositions at any one location do not exceed 3%. Hence, for a specific location, one calibration curve sufficed. If a different station is to be monitored, an analytically obtained calibration curve can be drawn from a theoretical model, provided that the composition of suspended solids in the new locality is known or determined.

Extensive tests were conducted to evaluate gage performance, including 1 week of flume tests at the Sedimentation Laboratory. The flume tests showed that the response of the gage is independent of flow rate in the range of 0 to 6 ft/sec and is capable of measuring sediment concentrations down to 500 ppm.

Further study on the effects of such influencing factors as temperature variation, electronic drifts, dead-time losses, compressibility effects, and statistical errors indicated that the induced errors were within the specified gage accuracy of ±20% at the lower concentration.

Arrangements are being made to construct a number of these units for independent testing by members of the Interagency Sediment Group at various locations in the United States.

Tracing the Movement of Beach Sediments

For many years coastal scientists and engineers have attempted to label sedimentary particles to determine their movement paths. Customarily these migrations are observed by adding dyed sediment or other foreign particles similar in nature to the beach sediments and then following their movements visually. Recently, however, improved methods have been developed for labeling natural or simulated sediments with radioisotopes as tracers. These new techniques permit more quantitative determination of sediment migrations.

The utilization of radioactive material as sediment tracers has increased in the past 10 years. After the initial test in the Thames River in England,[4] the use of this technique has spread until it is practically worldwide. Most of the objectives of these tests have been qualitative, e.g., using the tracer materials to determine movement path and sedimentation areas. Labeling techniques have varied widely and involve (1) plating or precipitating a thin film of radioactive material on the natural sediments, (2) using glass containing a radioactive tracer to simulate the natural sediments,[5] (3) incorporating of radioactive material within the natural elements or within simulated sediments, and (4) replacing by ion exchange some of the material sediment with a tracer material. Detection methods have also varied widely—Geiger-Mueller systems with one or several G-M tubes in gangs, scintillation systems making use of pulse-height spectrometry, and autoradiography. Sediment and tracer materials have been monitored *in situ*, or samples have been taken and measured in the laboratory.

In the spring of 1962, the U. S. Army Corps of Engineers and DID entered into a cooperative study to determine if the technique of using radioactive sand of the same particle size and density found at the Beach Erosion Test Site, Washington, D. C., would lead to more conclusive results than those obtained by other methods, such as the dye technique, in following migration. Fifty millicuries of ^{24}Na activity was induced by neutron irradiation in approximately 50 g of carefully prepared glass sand. The sand was distributed on the laboratory beach site

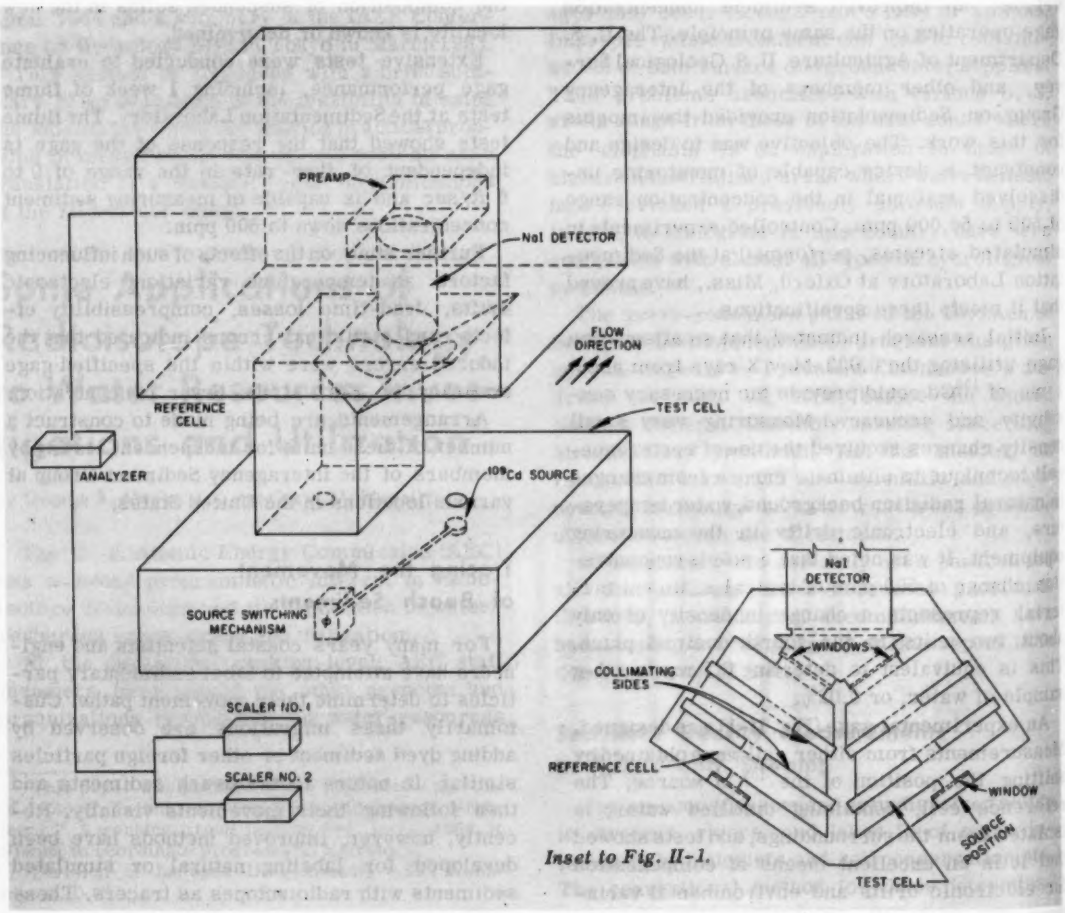


Fig. II-1 Experimental gage. Inset shows configuration of reference and test cell.

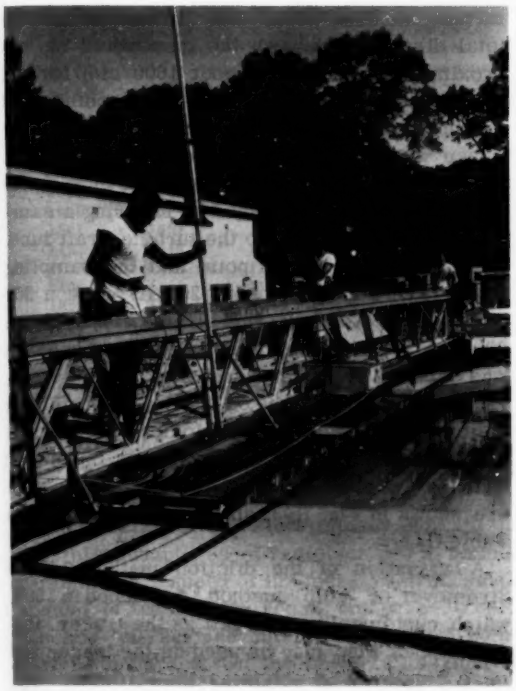


Fig. II-2 Measuring distribution of radioisotope-tagged sand at the U. S. Army Beach Erosion Test Site, Washington, D. C.

illustrated in Fig. II-2. The project was under the direction of Taney.[6]

The results of these tests were very informative. Data were taken by *in situ* measurement with NaI detectors and by autoradiographic measurements of core samples from various points on the simulated beach. These data permitted determination of the distribution pattern resulting from wave action and the depth of sediment penetration. Depth of penetration had not been determined previously in beach erosion studies.

The wave action was set for a 5-min period, followed by a search for tagged particles to determine distribution. At the end of the 5-min period, however, significant quantities of tracer had already reached the trap, which was located approximately 28 ft away. This indicated for the first time that the along-shore transport rate in the vicinity of the plunge zone is in excess of 5 ft/min. This rate of movement was not anticipated; previous studies using dyes indicated that wave action of approximately 30 min was necessary for measureable quantities of sand to reach the trap. Obviously the sedimentary particles move at velocities in excess of 5 ft/min. The wave-induced littoral current was known to be moving at a velocity of 45 to 50 ft/min. These studies have prompted the Beach Erosion Board to develop plans for use of radioisotopes in their future field investigations.

The feasibility of tagging sediments with the noble gas isotopes has very recently been demonstrated. Raising the temperature of a sediment sample in the presence of pressurized ^{85}Kr causes a useful quantity of the gas to diffuse into the sample and remain fixed. This appears to be an ideal tagging method because none of the sediment properties are modified. Initial studies are planned at the Beach Erosion Test Site.

Gaging Flow Through Turbines for Rating Purposes and Water Control

The power generated in hydroelectric turbines is one means of measuring the flow of water. If the turbine is properly calibrated, kilowatt-hours can be accurately related to water quantity.

The accuracy of water power turbine rating is important since this determination relates to the true measure of the water resources of an area and the available electrical energy. Accurate measurements are particularly important where rivers and streams cross state or national boundaries and where there are agreements concerning use of the water for irrigation purposes.

A number of methods are available for measuring discharge through turbines, but they have always proved expensive, time-consuming, or relatively inaccurate. For example, the well-known Gibson turbine-rating test measures the pressure surge in a turbine system in which water flow is suddenly stopped. A complex extrapolation of this increase in pressure reveals the amount of water flowing and the turbine efficiency. Another method, salt dilution,[8] requires special provisions to handle tons of salt.

A new and improved rating technique, developed in a joint program with the U. S. Geological Survey, has resulted in a several-fold increase in measurement accuracy through the use of ^{198}Au (Ref. 9). The experiments were carried out in two phases—one in the laboratory using a system of pipes and the other at turbine

installations of the TVA and of the Nashville District of the U. S. Army Corps of Engineers.

In each flow determination a known quantity of the ^{198}Au was added at a constant rate to the unknown flow, and its concentration downstream was determined by measuring the radioactivity either in discrete samples or in a continuous flow-through monitoring chamber. A scintillation counter was used to detect the tracer.

The results of the laboratory experiments showed the dilution method to be in close agreement with a standard volumetric method for determination of discharge from a pipe system. In a series of five tests, deviation from the dilution-method determination ranged from −2.5 to +1.8%, while average deviation was +0.2%.

For these turbine tests, injections of ^{198}Au were made into the water conduit or penstock and samples were obtained from the turbine-draft tube at Dale Hollow Dam in Tennessee. This installation has three turbines of approximately the same installed kilowatt capacity. During the time tracer experiments were being conducted, only two of the turbines were operated. Usually, three separate experiments were conducted consecutively at discharges of approximately 600, 1100, and 1600 cfs, beginning at the lower discharge rate. Comparable tail-water conditions were maintained for the studies by regulating the discharge through the turbine adjacent to the study turbine so that the total discharge through the dam would be approximately the same (about 1600 cfs) for all experiments. The penstock of the turbine used for the experiments was 15 ft in diameter; the intake was 80 to 100 ft below the lake surface. Tracer was injected at the penstock intake, and sampling was accomplished by lowering a sampling pump directly into the turbine-draft tube. Between the injection point and the sampling point (a distance of about 200 ft) were a 30° bend in the penstock, the turbine itself, and a 90° bend in the draft tube. Control discharge was obtained from a power plant flow integrator, rated initially in 1949 on the basis of Gibson calibrations and checked periodically since that time by current-meter measurements. Schematic drawings of the Dale Hollow Facility System are shown in Fig. II-3, and results are given in Table II-1.

A variation of the dilution method is the Stromeyer or "gulp" method,[8] developed in 1905, which customarily uses salt as the tracer. Radioisotopes also may be used as the tracer and were employed in obtaining the Stromeyer results listed in Table II-1 below. This total-count method of measuring discharge was perfected by Hull and Macomber[10] in 1958. In this method a known volume of tracer is injected rapidly into the stream; and, at a point suffi-

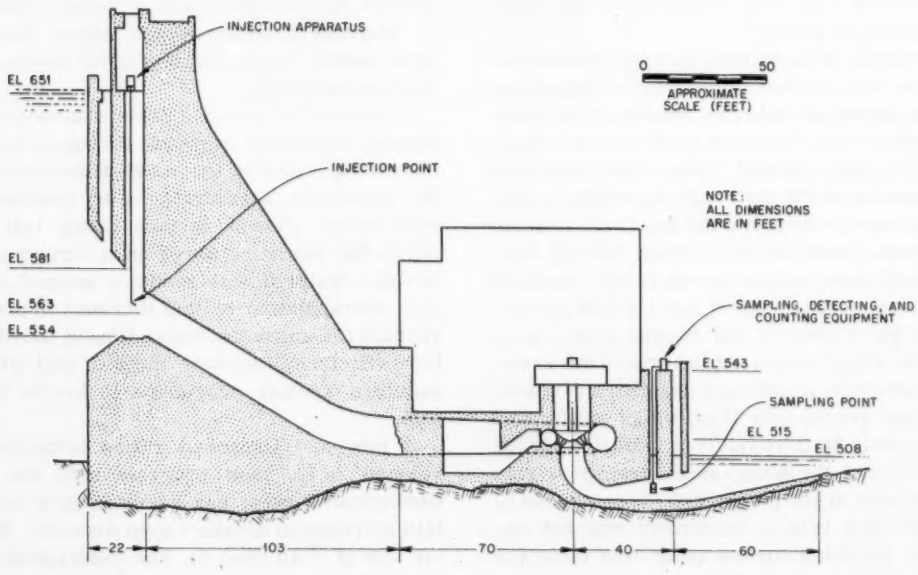


Fig. II-3 Cross-section through penstock and draft tube on one unit.

Table II-1 COMPARISON OF COMPUTED TO CONTROL WATER DISCHARGE IN FIELD EXPERIMENTS[11]

Control discharge, cfs	Computed discharge, cfs		Difference computed from control, %	
	Dilution method	Stromeyer method	Dilution method	Stromeyer method
1600	1543		−3.6	
1639	1680	1658	2.5	1.2
1650	1574		−4.6	
1661	1651	1625	−0.6	−2.2
1153	1146	1170	−0.6	1.5
1150	1112	1154	3.3	0.3
1104	1155	1116	4.6	1.1
1592	1590	1543	−0.1	−3.1

ciently far downstream for complete mixing to occur, measurements of the tracer concentration are made. A graph of concentration vs. time is plotted, and the average concentration is determined from the graph.

The results of this study clearly indicate the possible advantages of radioisotopes in turbine ratings, e.g., speed of the measurement, small amount of tracer required as compared to salt, absence of service interruptions, and comparable accuracy. On the basis of information derived from these low-head turbine studies, the Bureau of Reclamation plans to investigate the adaptation of the method to high-head turbines.

Measurement of Water Flow in Porous Media

Groundwater hydrology has never been fully understood because of the many complex factors, such as the nature of soil, water-flow gradient, porosity, and particle size. The better understanding of these parameters should give insight to the distribution of subsoil water and the discharge of man-made wastes. Such knowledge should aid in our achieving better water supplies, particularly along salt-intruded coastal areas.

Conventional methodology has relied on moisture determination or on conductivity-measurement techniques to trace saltwater intrusion. These methods have a number of limitations and usually supply only qualitative data. For example, in collecting samples for moisture determination, the physical disturbances upset the moisture equilibrium. Conductivity determinations of saltwater intrusion involve a source of error since they are measured at finite points. In addition, profiles are difficult to obtain.

The U. S. Geological Survey's Phoenix Laboratory has undertaken several tasks to determine the migration of moisture in porous materials and the mass transport of salt water into aquifers. These problems are being investigated by means of hydraulic models and mathematical analysis. Radiotracers in the hydraulic models have proved especially well suited for laboratory analysis. The following projects are now under way:

COASTAL SEA-FRONT MODEL

A hydraulic model 23 ft long, 3 in. thick, and 18 in. high, simulating conditions in a shallow aquifer along a profile transverse to a seafront was constructed to enable study of diffusion near the interface between salt water and freshwater. Silica sand was used as the porous medium.

Radiotracers (^{32}P and ^{131}I) were used to define the band of dispersion between freshwater and salt water resulting from the reciprocating motion of the water caused by tides.

This hydraulic model was patterned from field data collected at the Biscayne aquifer in the area of Miami Beach, Fla. However, the analysis can be readily applied to other regions. The Biscayne aquifer was chosen because a large quantity of data has been compiled on its behavior.

Studies on the operation of the Biscayne model have revealed the adequacy of present mathematical theory describing the dynamics of flow taking place in the region of a saltwater-freshwater interface.

RATE OF FLUSHING MINERALIZED WATER FROM A GENERALIZED AQUIFER

One problem that confronts the groundwater hydrologist is determining the rate of flushing of mineralized waterlogged aquifers. In irrigation practice the surface is flooded with freshwater, and the mineralized water is removed from the aquifer by pumped wells. The wells discharge into canals that are lined to cut down on seepage loss and to transport the drainage water away from the area. It is desired to know the time required to dilute the initial mineral-laden water in the aquifer to an acceptable level.

In the past the problem has been studied with the use of different colored dyes in models, but the dyes have proved inadequate for complete analysis of sections removed from the models.

The U. S. Geological Survey has built a hydraulic sand model for these studies. The

model simulates the actual prototype field lithology as well as the rates of pumping and recharge and therefore will give some indication concerning the rates of dilution in actual field regions.

Simulated irrigation water containing a radioactive tracer is applied to the surface of the model while it is being pumped. Samples of effluent water are collected at predetermined intervals and counted. From the time and concentration of the tracer, dilution and recharge rates can be determined.

Radioisotopes have the advantage of being easily detectable in cross-sectional slices by autoradiography and thus give some indication of the degree of adsorption of the tracer in the model. Any appreciable adsorption of tracer will bias the measurements unless corrections are applied. Phosphorus-32 was used as a tracer in these studies; although more complete data were obtained than with dyes, which were also used, the results suggested that the ideal technique might involve use of less readily adsorbed radiotracer, such as ^{131}I. The results from the reruns using ^{131}I are now being evaluated. Preliminary data from these studies already have been used by Skibitzke to analyze water-flow problems in Pakistan, Mexico, and Southern California.[12]

DETERMINING DIRECTIONAL PERMEABILITY IN POROUS MEDIA

The study of migration of soil moisture in both unsaturated and saturated media is made difficult by the problem of measuring the directional characteristics of permeability, i.e., the permeability tensor. A radioactive tracer was used to define the directional permeability of saturated isotropic and anisotropic porous media. An isotropic medium represents an aquifer in which the sand grains are homogeneously dispersed and an anisotropic porous medium, an aquifer in which the sand grains are of different shapes and sizes and contain some fissures. Water flow in the isotropic medium should be uniform without streaming. In the anisotropic medium, streaming effects are common around large grains and along fissures. Two models were constructed: an isotropic one of uniform sand cemented with epoxy resin; and an anisotropic one cut from a block of Carrizo sandstone from Austin, Tex. Both models were slowly saturated with distilled water to allow the rock interstices to become saturated by capillary action. Saturating the block required more than 14 weeks. Then about 5 ml of ^{32}P solution was injected into a borehole in the center of each model. When the tracer had spread through the rock interstices, the excess moisture was drained and the blocks were dried.

The blocks were sawed into horizontal sections for analysis. Autoradiographs revealed the extent of migration of the tracer. The isotropic medium showed uniform migration in both direction and magnitude. The anisotropic medium showed appreciable directional migration, principally because of a small fracture. Other types of anisotropic media are being studied.

SOIL MOISTURE-VAPOR INTERFACE

The movement of moisture through unsaturated soil may occur as liquid or vapor and may proceed upward or downward. Studies to establish more fully the dynamics of such movements are under way at the Phoenix Laboratory.

A large cylindrical tank 24 in. in diameter by 72 in. high was filled with uniform-grained sand. A 1-in.-diameter well of thin-walled aluminum pipe was inserted into the center of the sand column. Initially a neutron-scattering probe was moved vertically through the soil column as moisture was applied on the surface. The neutron probe did not prove satisfactory, and a radioisotope thus was used to follow migration of the water to the site where it was transformed from liquid to vapor. The radiotracer was introduced into the water at the saturated bottom of the soil column, and the movement of the water was monitored as it migrated upward to the surface or to the point where movement ceased. Data from such tests are useful in determining the rate of evaporation from land surfaces near rivers.

DETERMINATION OF THE VOLUME OF UNDERGROUND WATER RESERVOIRS

In arid and semiarid regions, underground water storage is a better method than surface storage because there is little or no loss by evaporation. The main disadvantage is assessment of the quantity stored.

In past studies, conventional tracer methods, such as fluorescein dyes, salt solutions, and variations in chemical composition of the water have given approximate data on storage capacity. Successful use depends largely on the type of

aquifer since the tracer may be adsorbed or precipitated, with a resulting error in the measurement.

The radioisotope ^{3}H should be an effective water tracer. It can be converted into tritiated water, which undergoes all the physical and chemical changes normally associated with water; and, although it is difficult to detect and measure in the field, it can be measured with radiation counters in the laboratory.

The U. S. Geological Survey has used tritium to determine the capacity of the Lake McMillan Underground Reservoir in Eddy County, N. Mex. This site was chosen because of previous data available from earlier dye and chloride studies. These data indicated that water leaks through the floor of the lake to underground reservoirs. Lake McMillan is undergirded with a gypsiferous formation that permits considerable leakage. This formation contains solution channels and large caverns. From these channels and caverns, the water emerges at Major Johnson Springs about $3\frac{1}{2}$ miles downstream.

In April 1961 the lake was estimated to contain about 17,400 acre-ft of water. Previous determinations on the radiation background content of the surface water showed the lake to have a tritium concentration from 40 to 50 tritium units (T.U.), where 1 T.U. = 1 atom $^{3}H/10^{18}$ atoms ^{1}H; 150 curies of HTO was added to raise the concentration to about 2200 T.U. (7.12×10^{-6} $\mu c/ml$).

Samples from the Major Johnson Springs area were collected and analyzed. Peaks in ^{3}H concentrations indicated that the travel time of the HTO from Lake McMillan to the sampling point was about $2\frac{3}{4}$ months, which is comparable to the travel time of 2 to 3 months determined by the chloride technique.

This represents a probable minimum travel time for a significant quantity of water to pass from the lake to the springs. The results from the ^{3}H study indicated an underground water system having a volume of approximately 50,000 acre-ft, an amount that compared favorably with data from previous studies.

Precise evaluation of the ^{3}H data was hampered by the short residence time of the ^{3}H in the lake and the ^{3}H "rain-out" from nuclear testing in late 1961. However, this development has demonstrated the usefulness of a valuable new tool for perhaps a better solution of a puzzling hydrological problem. A report on the study with ^{3}H is available.[13]

Determining Fluid Dynamics of Repressured Petroleum Reservoirs

Many petroleum reservoirs must be periodically repressurized with water to maintain oil production. Standard practice calls for pumping water into input wells and forcing the oil toward the production wells. However, the distribution of the oil and water in the formation is difficult to know without the use of tracers. Considerable progress has been made in adapting various radioisotopes to meet this problem.

A cooperative project for developing underground isotope tracer techniques is now under way with the Bureau of Mines, Petroleum Research Center, Bartlesville, Okla. The information derived from these techniques is applicable not only to the petroleum recovery process but to many water-tracing problems.

Two major field experiments, together with a number of confirming laboratory experiments, have been conducted to determine the feasibility of using radioactive tracers, primarily ^{3}H, in the evaluation of miscible-phase petroleum recovery operations. The fluids traced during this work were *n*-heptane as a component of gasoline, isopropanol, and water.[14] In one field experiment the relative vertical distribution, as defined by core saturations, was determined by drilling a second well to the producing horizon near the water input well and coring the formation of this second well. Analysis of the cores and the extracted fluids provided data from which a curve was plotted showing vertical distribution of the injected liquid. In hydrology studies the technique can be used to determine which formation is supplying water and the location of fissures that permit contamination of an aquifer.

Another noteworthy field experiment was conducted to determine the feasibility of using ^{85}Kr as a water tracer in underground porous medium. Krypton-85 is particularly attractive as a water tracer because it does not enter into chemical or metabolic reactions, has a 10.6-year half-life and emits an easily detectable 0.7-Mev beta radiation.

The field test using ^{85}Kr was preceded by a series of laboratory tests with consolidated core flow systems. These laboratory tests showed considerable promise. Previous efforts in this area had failed because of the difficulty of removing the ^{85}Kr from solution for counting by the standard gas-counting procedures. The

problem was overcome in this series of tests by utilizing a scintillation dip counter. Although some ^{85}Kr is lost from solution during the interval between sampling and counting, a sufficient amount remains to permit qualitative analysis.

Despite the promising laboratory tests, field studies disclosed considerable difficulty with the solubility of ^{85}Kr. In hydrocarbons it is about tenfold greater than in water. Consequently, when a water solution is in contact with hydrocarbons for extended periods, as occurs in petroleum reservoir tracer work, ^{85}Kr has marginal usefulness as an oil field tracer. However, the technique may be quite useful as a water tracer in aquifers that remain under relatively constant pressure with no open flow. Reports on the use of ^{3}H and ^{85}Kr for reservoir tracing are now being prepared.

Tritium Dating

Prior to the discovery that tritium could be used to determine the age of relatively recent rainwater, there was no method for performing this analysis.

In a cooperative program between AEC and the Internal Revenue Service, ^{3}H-dating techniques were developed to determine the age of brandy and other distilled spirits.[15] The information is used in determining tax rates on alcoholic beverages. The Internal Revenue Service has applied identical techniques to determine the recharge rate of aquifers for tax purposes.

A tax case tried in Lubbock, Tex., in January 1962, involved a question of water depletion from the Ogallala water-bearing formation. The issue was cost depletion allowance for water pumped for irrigation purposes. The plaintiff claimed that recharge of the aquifer was less than the water pumped out; hence the aquifer would soon become depleted. The government contended that the aquifer was getting sufficiently recharged and had ^{3}H data to prove the point. The case is still in court.

The second court case, in November 1962, also involved water in the state of Texas. The plaintiff was seeking long-term capital gain on water being sold to the Great Northern Natural Gas Company. The government contended that the proceeds from the sale of this water were income and should be taxed as income instead of capital gain, because the plaintiff did not make an outright sale of his wells. The government also claimed that the wells contained ^{3}H at the 800-ft level and were getting constant recharge from rainfall. The government won the case in February 1964.

Measurement of Snow Density Profiles for Water Management

In California some 300 mountain snow courses are measured regularly throughout the winter. The information is used in forecasting the water available for power, domestic water supply, and agriculture. Work on radio-snow gages has been reported in a recent issue of this review[2] and is not repeated here. The use of these probes promises a superior nondestructive method of measuring the water equivalent in snowpacks, including water in transit through the pack or ponded on the soil, or frozen as ice layers or lenses within the pack. In addition, tests indicate that the radioactive sources could be used to evaluate the thermal quality of the snowpack and to determine the amount of heat required to melt the pack.

Deep-Water Isotopic Current Analyzer (DWICA)

One of the many problems confronting both the hydrologist and the oceanographer has been the lack of a suitable method to measure precisely and record accurately direction and flow of slow-moving currents in the range of 0.01 to 1 knot.

Conventional methods, such as the Savonius type, fail to give reliable results due to the mechanical friction drag on the system which occurs in the case of a rotor mounted on a sapphire bearing and the existence of different response-time constants for acceleration and deceleration.

A new method developed by the William H. Johnston Laboratories is based on an isotopic principle. The technique depends on injection of a radioisotope into a moving-water environment and measuring drift as the radioactivity passes a detector. This system has also been described in detail in a previous issue of this review.[1]

Conclusion

Numerous lines of research have been pursued to increase the actual use of isotope

techniques by scientists and engineers engaged in the study of water resources. Much of the technology is already in use assisting in the investigation of complex hydrological action and helping to solve everyday water-resources problems.

The AEC has sponsored a special study of the potential isotope techniques in hydrological research.[16] Applications range from those obviously feasible to others with only a small probability for success. The list of potential areas of study is periodically reevaluated in light of new advances in materials, techniques, and instrumentation. There is a waiting list of good proposals in this important public-benefit area that warrant further exploration as funds become available.

Review of the Tokyo Conference

A symposium on the use of radioisotopes in hydrology, organized by the International Atomic Energy Agency, was held in Tokyo, March 1963, with the assistance and cooperation of the Japanese govenment. It was attended by about 100 participants from 14 countries: Australia, France, the Federal Republic of Germany, Indonesia, Iran, Israel, Italy, Japan, the Netherlands, the Philippines, Sweden, Thailand, the United Kingdom, and the United States of America. There were also participants from 5 international organizations: the Economic Commission for Asia and the Far East (ECAFE), the Food and Agriculture Organization (FAO), the International Union of Geodesy and Geophysics (IUGG), the World Meteorological Organization (WMO), and the International Atomic Energy Agency (IAEA).

Twenty-seven papers with discussions and abstracts are presented in the published proceedings of the symposium.[17] The applications of radioisotopes to the solution of some of the general problems facing the hydrologist today are aptly stated in the following portion of the Foreword:

> The increasing emphasis on the development of water resources poses problems which are of interest to all countries, both developing and advanced, where the demand for water is continuously rising. There is no doubt that greater efforts must be made to evaluate, control, and develop water resources using all scientific means available and during recent years increasing attention has been directed to the supplementation of hydrological methods with radioisotope techniques. These techniques have already been applied to a number of problems and their potential usefulness demonstrated.
>
> Radioisotopes can be used for stream discharge measurements with an accuracy as good as that obtainable with conventional methods. They are also finding increasing application in the measurement of groundwater direction and velocity, the study of suspected interconnections between different sources of water, and the investigation of mixing processes in rivers and lakes. Radioisotope techniques have been used in different parts of the world for studying the transport of silt in rivers and harbors. Present research is directed toward making these investigations on a quantitative basis which, if successful, would be of great importance in the design of hydraulic structures.
>
> The method of finding out the age of groundwater by measuring its natural tritium content can be applied to the determination of the recharge rate of groundwater bodies, so enabling a more rational use of the groundwater reserves without fear of overexploitation. Current research is aimed at using carbon-14 for groundwater dating to extend the age measurable by tritium.

Papers were presented at the symposium on these various aspects of hydrology. In the present review the selection of radiotracers and their specific applications are described.

Selection of Tracers

The applications of several different tracers, some in combination, as well as the use of natural isotopes, are described in the various papers. The choice of a tracer for a specific problem is governed by several factors. The tracer should be stable and soluble in water, have a half-life suitable for the type of problem for which it is to be used, emit radiation that is easily measurable in the field where possible, have sufficient specific activity, and be reasonable in cost. It should not exhibit any appreciable adsorption within the test environment or contaminate the drinking waters above the maximum permissible level.*[18]

TRITIUM

^3H (12.26 Years, 0.018-Mev β, No γ). Tritium, which was used more frequently than any other isotope, has several advantages as a tracer (HTO); it is not readily adsorbed, is obtainable with high specific activity, is one of

*References 18 to 44 are listed in the order of their appearance in the published proceedings (as corrected by the errata sheet) rather than in the order of their appearance in this review.

the least toxic isotopes, and is inexpensive.[18] However, because of its soft radiation, tritium cannot be detected and measured conveniently in the field. Natural tritium produced by cosmic radiation is the most useful isotope for groundwater dating, since it labels rainwater with a tritium content characteristic of the locality. However, its use as a dating tool has been curtailed by thermonuclear testing, which adds to the natural tritium level. In one instance, bomb-produced tritium from rainwater was used as a tracer for groundwater movement at Carrizozo, N. Mex.[27,45] Also to be considered is the fact that its use as an artificial tracer may prevent the employment of naturally occurring tritium in hydrological problems for several years to come.[35]

HALIDES

^{131}I (8.05 Days, 0.34- and 0.61-Mev β's, 0.36- and 0.64-Mev γ's);
^{132}I (2.3 Hr, 1.5 to 2.1-Mev β's, 0.53 to 0.96-Mev γ's);
^{82}Br (35.9 Hr, 0.44-Mev β, 0.55 to 0.78-Mev γ's);
^{36}Cl (3×10^5 Years, 0.71-Mev β, No γ). The use of isotopes of bromine, iodine, and, to a lesser extent, chlorine as tracers is described or mentioned in a number of the papers (see Refs. 18, 19, 22–24, 27, 32, 33, and 36–38). These anions are not adsorbed to a great extent and are therefore generally suitable as tracers.[18,37] To reduce the chance of adsorption of the tracer, a quantity of natural carrier can be added.[22,23] If field measurements are inconvenient, iodine tracer can be precipitated as silver iodide, with a small amount of natural iodine added as a carrier, and counted in the laboratory.[23,38]

Iodine-131 and ^{82}Br, with half-lives of 8 days and 36 hr, respectively, are not suitable for experiments of long duration. Iodine-132, which can be obtained by elution from a column containing ^{132}Te, can be used for experiments of very short duration[18,22] and allows consecutive experiments in coastal areas without interferences from injections at the time of the previous tide.[22]

The chloride ion ($^{36}Cl^-$) appears to be useful as a tracer but is limited by its high cost, long half-life (3×10^5 years), and pure beta radiation which is difficult to detect and measure in the field.[37]

SODIUM

^{24}Na (15.0 Hr, 1.39-Mev β, 1.37- and 2.75-Mev γ's). Since most streams and rivers contain at least trace amounts of natural sodium, ^{24}Na can be used as a tracer without too much loss by adsorption, especially over relatively short distances. However, some losses over longer distances (several hundred meters) have been detected.[18]

Sodium-24, which has a half-life of 15 hr and emits hard gamma as well as beta radiations, was used frequently as a tracer (Refs. 18, 21, 23, 24, and 44). Although the gamma radiation may present problems in transportation and shielding, it has the advantage of direct measurement in the field. Samples of the streams containing ^{24}Na tracer were also taken to the laboratory and analyzed by separation of sodium with an ion-exchange column and subsequent beta counting.[23] Sodium-24 was also used in tracer experiments to measure sand transport. The applicability of glass particles as a sand tracer was determined by neutron-irradiating sodium glass particles of the proper size and density to produce ^{24}Na for comparison studies with irradiated natural sand.[24]

PHOSPHORUS

^{32}P (14.3 Days, 1.71-Mev β, No γ). The use of ^{32}P as a tracer was generally unsatisfactory because it is subject to adsorption.[18,32,38] In one paper it was reported that tracing of ^{32}P as phosphate was restricted in groundwater containing Fe^{3+} and Al^{3+} and impossible when the *p*H was greater than 3.5 (Ref. 38). Phosphorus in phosphate form may also be absorbed as a nutrient by aquatic weeds or animal life.

STRONTIUM

^{89}Sr (51 Days, 1.46-Mev β, Essentially No γ); ^{90}Sr (28 Years, 0.54-Mev β, No γ; also 2.27-Mev β from ^{90}Y Daughter). None of the papers described the use of ^{90}Sr alone as a tracer; however, three papers[34,42,43] report using ^{90}Sr with another isotope. It is interesting to note that some of these studies employed strontium of fallout origin.[42,43]

CARBON

^{14}C ($\sim 5.7 \times 10^3$ Years, 0.16-Mev β, No γ). The only application of the naturally occurring ^{14}C isotope was for the determination of groundwater ages.[39,40] Carbon-14 as bicarbonate in

water can in some cases be used to determine residence time up to 40,000 years.[39] The ratio of ^{13}C to ^{12}C was used to study isotopic exchange which could affect the accuracy of ^{14}C dating.[39]

OTHER ISOTOPES

For some hydrological studies it is desirable to have isotopes of longer half-lives than those of ^{24}Na, ^{82}Br, and ^{131}I and yet not as long as that of tritium. In the form of stable complexes, tracers are less subject to adsorption and exchange; thus radioisotope complexes that meet the longer half-life requirement may be useful as tracers. Examples of such complexes are the ethylenediaminetetraacetic acid (EDTA) chelates of ^{46}Sc, ^{51}Cr, ^{114m}In, and ^{124}Sb (Ref. 37).

^{51}Cr (27.8 Days, No β, 0.32-Mev γ). Chromium-51 as the EDTA complex appears to have several advantages. It has no beta activity, its gamma activity is of low energy, and its permissible limit in drinking water is therefore relatively high. In double-tracer experiments with tritium, it was demonstrated that this chromium complex was transported as rapidly as tritium water, even in concentrations of 0.01 ppm.[37]

^{106}Ru (1.00 Year, 0.04-Mev β, No γ; also 2.0- to 3.5-Mev β's, 0.51- to 2.66-Mev γ's from ^{106}Rh Daughter). Another complex described as a possible tracer was nitronitrosylruthenium. Ruthenium-106 has a half-life of 1 year and is therefore in a range where there are few radiotracers. Although there is a variety of ^{106}Ru complexes, the nitro complexes are among the most stable. Percolation tests were made on columns of aquiferous sand and very argillaceous soil using this tracer.[30]

^{60}Co (5.27 Years, 0.31-Mev β, 1.17- and 1.33-Mev γ's);

^{65}Zn (245 Days, 0.33-Mev β+, 1.11-Mev γ, Electron Capture);

^{46}Sc (84.0 Days, 0.36-Mev β, 0.89- and 1.12-Mev γ). Cobalt-60, ^{65}Zn, and more recently ^{46}Sc have been used to prepare radioactive glass sand in experiments on littoral drift.[26] The gamma activity of these isotopes made possible the measurement of radioactivity on the seabed for tracing the sand movement.

^{198}Au (2.70 Days, 0.96-Mev β, 0.41-Mev γ). Gold-198 was used as a tracer in open-channel flow measurements. There was some indication that aquatic weeds may absorb and/or adsorb this radioisotope.[20]

^{86}Rb (18.6 Days, 0.72- and 1.77-Mev β's, 1.08-Mev γ). Rubidium-86 chloride was used with ^{32}P in studying the movement of acid hot-spring water. Losses of both tracers by adsorption were indicated.[38]

Specific Applications of Tracers

The most frequent tracer applications described were related to the measurement and study of surface and groundwater movements. Tracers were also employed to measure and follow the transport of silt and sand in both laboratory and field studies. Other applications included dating studies and determination of meteoric runoff. Two papers describe the contribution and application of radioactive tracers in groundwater hydrological studies and research.[27,29]

FLOW MEASUREMENT OF SURFACE WATERS

The dilution, continuous-sample, and total-count radioisotope methods of flow measurement were described and used in a comparison study by Clayton and Smith[18] on three rivers of different geological character. All three methods, which are accepted as providing accurate flow measurements in closed conduits, may be satisfactorily used; however, the authors preferred the continuous-sample method with gamma-emitting tracers since they found it provided operational convenience, flexibility, and the highest accuracy with the least amount of tracer. The study indicated that ^{24}Na, ^{82}Br, and ^{3}H may be used reliably as tracers; however, ^{3}H presents difficulties in that it can neither be measured nor detected directly in the field. Phosphorus-32, when used without a carrier, was unsatisfactory because of high adsorption.

Guizerix et al.[19] measured discharges of rivers and hydroelectric penstocks by the total-count method. By sampling a portion of the flow with gages and constant-level tanks during passage of the ^{82}Br tracer, they were able to count both the sample and an aliquot part of the tracer solution in the same geometry. This method avoided the difficulties of counting geometry, made possible the determination of statistical counting accuracy, and gave an indication of mixing efficiency.

Timblin and Peterka[20] describe open-channel flow measurements using ^{198}Au. They conclude that with the development of more precise equipment and procedures, radioisotope methods of flow measurements will be used in many field applications, including rating of hydraulic structures, calibration of gaging stations, and measurement of seepage losses from canals. Minimum mixing distance must be determined and isotope losses must be either minimized or taken into account to achieve these objectives.[20]

Kato et al.,[23] in experiments on the Sorachi River in Japan, used ^{24}Na and ^{132}I to obtain precise information on the flow times and flow conditions of water to be released from a multipurpose dam under construction. They found that the maximum tracing distance at one time, with salt water added as tracer, was about 4 km, while with radiotracers it was as great as 30 km. The accuracy of measurement with radiotracer in a 10-km distance was about equal to that for 2 km with salt water. For the detection of the activity, both periodic collections of 1-liter samples for subsequent analyses and direct measurements with the underwater detectors were made for purposes of comparison. In the laboratory the tracers were concentrated with natural carriers by precipitating the ^{131}I as AgI and recovering the ^{24}Na from an ion-exchange column. The beta activities of the tracers were determined; two flow curves, the first obtained by this method and the second by the rate meter readings, were compared.

A method for measuring velocity and dilution rate in a river with both a radioactive $^{24}NaCl$ solution, injected as a point source, and nonactive NaBr or NH_4Br, injected continuously, is described by Kobayashi and Nukazawa.[44] The radioactive tracer solution was poured into the water at the beginning of the continuous injection, and scintillation detectors that were placed downstream measured directly the changes in activity and indicated the arrival of the nonactive tracer. Samples were taken at regular intervals for laboratory determination of both the ^{24}Na activity by counting and concentration of the nonactive tracer by activation analysis (^{24}Na, ^{80}Br, and ^{82}Br). Dilution rates were calculated by both methods. This technique permits the tracing of a continuously injected nonactive tracer using a minimum amount of radioactivity.

GROUNDWATER STUDIES

Harpaz et al.[27] reviewed the place of isotope methods in groundwater research. The paper presents a classification of isotope methods, which are suitable for comparison with accepted hydrological methods. In certain cases isotope techniques not only replace and improve existing methods, but they may provide immediate answers to some hydrological problems. Several authors have given descriptions of evaluations of isotope methods grouped according to the isotope used; however, to evaluate the relative merits of isotope and conventional hydrological methods, Harpaz et al.[27] suggest a different classification, i.e., dating, pulse tracing, and isotopic analysis.

Distinctive features of isotopes as tracers are:

1. The wide variety of isotopes of different chemical and radioactive properties which are available and the sensitive detection methods which can be used for their identification.
2. The inherent time factor of decay associated with the radioisotope; a single measurement with isotopes can replace extended periods of hydrological observations.
3. The existence of tritium, deuterium, and ^{18}O, which are isotopes of the elements in water and hence ideal tracers for the unperturbed identification of bodies of water.
4. The change in stable isotopic abundance ratios by the basic physicochemical process of the natural water cycle, such as condensation and evaporation, which results in a unique characterization of water from different sources.

The flexibility and relative ease of obtaining isotopic data provide short-cut methods for the solution of some general hydrological problems; however, relative to conventional methods, these are limited in their contribution to the determination of hydraulic parameters. Nevertheless, all characteristics related to water movement are most directly measured by tracing methods in general and by isotope methods in particular. Characteristics related to actual travel path of individual water particles are evaluated only with isotopes of elements in water.

The hydrological phenomena affecting the use of tracers in timing groundwater flow are discussed in a paper by Theis.[28] He points out that the determination of the amount and character of field dispersion and the nature of

movement through the zone of aeration to the water table are two fields of investigation that appear particularly suitable for the use of tracers. Further efforts in these fields are necessary for intelligent application of tracers to groundwater studies. Tracers also lend a direct approach to the problems of movements of contaminants in groundwater; however, they cannot serve as a shortcut in the absence of other hydrological data.

In the investigation of an underground semitidal and partly saline pool, Smith and Clark[22] determined the general direction of flow of currents during flooding and ebbing tide with ^{132}I and a buoyant "spider" containing six waterproof Geiger counters. By using ^{131}I they estimated pool volume, the volume in the whole pool system, the volume entering the pool during flood tide, and the throughput of freshwater.

Moser et al.[33] describe the measurement of flow velocity, both across a filter tube and vertically. The vertical flow rate as well as the affluents and effluents are of prime importance to hydrologists, and determination of these values is possible under certain conditions. The technique involved the use of ^{131}I and ^{82}Br, with multiple detection instruments suspended in the filter tube above and below the point of injection (Fig. II-4).

Iwai and Inoue[34] propose a method for the estimation of groundwater stratification. After determining groundwater velocity and its vertical distribution by the point dilution method, one could inject tritiated water and a cation such as ^{90}Sr into a well and determine their arrival times at a second well. By comparison of the ratio of the two arrival times and by certain other calculations, the stratification of groundwater near the two wells may be estimated.

Burdon et al.[35] used tritium to trace groundwater in a limestone region (karst) in Greece to determine possible interconnections between two sinkholes and coastal springs. For utilization of the groundwater and possible interception of that being lost to sea, information was required on the following three points:

1. Which sinkhole feeds which spring or group of springs?
2. What is the groundwater residence time between the sinkhole and the springs?
3. How much groundwater is stored between the sinkholes and the springs?

The existence of two interconnections was demonstrated, and results were applied to the determination of transit times, flow rates, and groundwater storage.

Degot et al.[36] describe two uses of ^{82}Br. In one, labeled NH_4Br was injected into a central shaft, and then the arrival of the activity in a neighboring shaft was surveyed. The results

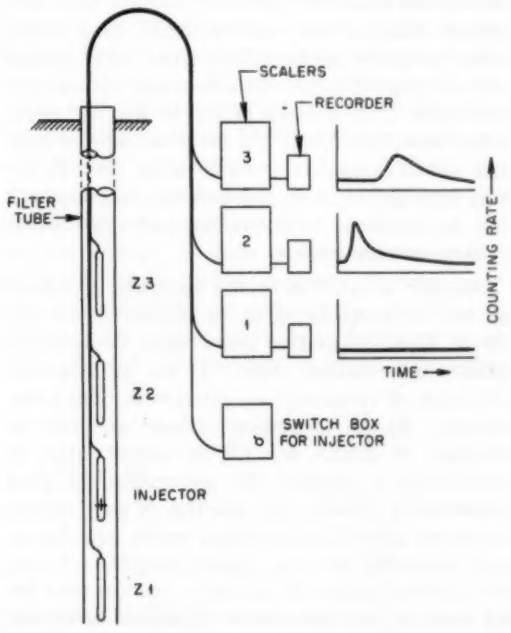


Fig. II-4 Setup for the determination of vertical flow in filter tubes.[33]

provided clarification of distribution velocities in an alluvial formation, and they represent a contribution to the verification of Theis's law under conditions which depart from the basic assumption. In the other, ^{82}Br was used in the localization of permeable strata or seams in a geological calcareous marl series intended to support quay walls in a harbor basin. The tracer was forced into the formation with inactive water; the water was then pumped back and the recovered fraction of the tracer estimated. Localized as well as overall activity measurements permitted precise location and profiles of the zones of recharge. The authors emphasize that tracers represented the only known means for solution of these problems.

TRANSPORT OF SAND AND SEDIMENT

Radioactive tracers have been used for about 10 years in the study of sediment movement. The results, however, primarily qualitative, give the direction and speed of movement but not the volume of sediment transported over a certain time. Lean and Crickmore[24] discuss four methods by which quantitative results may be attained in the case of unidirectional sand transport: space-integration, time-integration, steady-dilution, and cloud-velocity. They tested some of these methods in a laboratory channel with a rippled sand bed containing radioactive particles. The authors conclude that the problems associated with the measurement of sand and silt transport in rivers using tracers are still formidable, and, although the four methods can be applied, each has its particular difficulties and application.

Chabert et al.[25] describe radioactive tracer studies of movements of sediments in rivers. Their first qualitative tests were designed to gather information about (1) the development of zones of transport in relation to liquid discharge, (2) the formation of bars and rate of advance of dunes, and (3) the possibilities of correcting a channel for navigation. In later quantitative studies the labeling of an alluvium of coarse gravel and pebbles, which was classified according to size, made possible not only the determination of hydraulic parameters for the onset of entrainment as a function of diameter and derivation of transport granulometry as a function of discharge, but also permitted calculation of the sediment discharge rate by a transport formula. Adaptations of dilution methods (continuous dilution and total count) to the measurement of sediment transport were also being studied.

Radiotracer experiments on littoral drift in Japan are reported in a paper by Kato et al.[26] Glass sand tracers containing either ^{60}Co or ^{46}Sc were used. The tracers were prepared with the desired specific gravity by adding cobalt to the ^{60}Co and lead to the ^{46}Sc glasses during preparation and then grinding the glasses to the grain size distribution specified for the particular place of observation. The methods and equipment for depositing the tracer, the kinds and characteristics of the underwater detectors, some considerations of radiation protection, and incidental tracer techniques are also described. Figure II-5 shows one of several pieces of depositing equipment. Radioactive glass sand and soap are placed in the chamber shielded with lead walls and sealed at the bottom with vinyl sheet. The equipment is lowered from a boat, and a plumb weight is dropped along the messenger wire, activating the release mechanism. The annular cutter pierces the vinyl sheet, and the spring-driven piston forces out the radioactive glass sand.

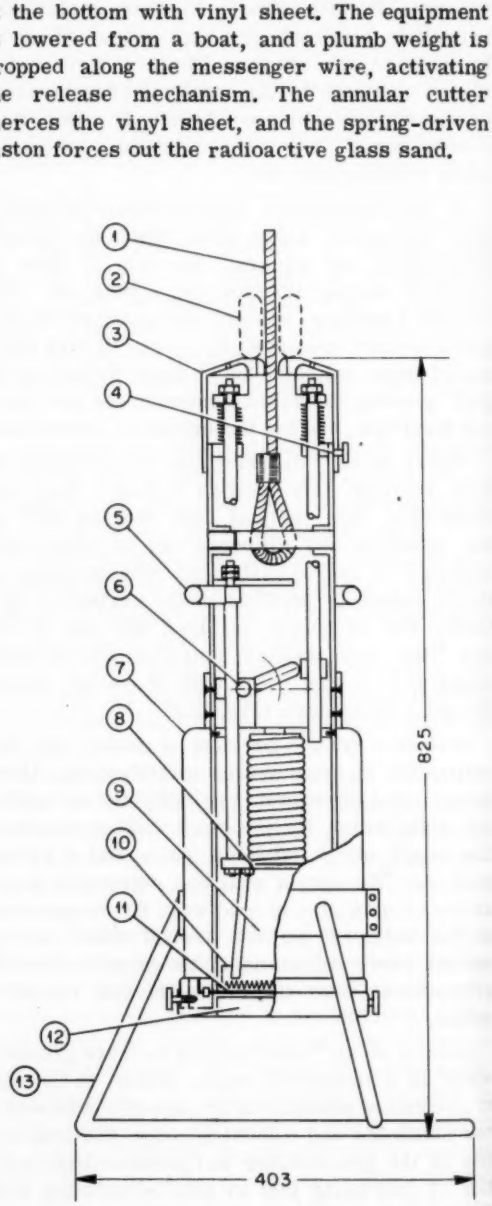


Fig. II-5 Depositing equipment used at Tomakomai.[26] Dimensions are in millimeters. (1) Messenger wire, (2) hammering plumb, (3) cap, (4) clamping screw, (5) handle to compress spring, (6) claw hook, (7) shielding lead, (8) piston, (9) charging space for radioactive glass sand, (10) annular cutter, (11) vinyl sheet, (12) leaden cover for shielding, and (13) stand.

DATING

In conventional tracing of groundwater movement with radioisotopes, the tracer is injected at one location and the time of its arrival is determined at another. However, in the situation where movement is slow enough that the tracer would not appear for tens, hundreds, or thousands of years, the use of artificial injection is obviously unacceptable. By comparing the measured activity of a natural radiotracer (for example, ^{14}C) in the water being studied with the activity of the same isotope in surface water, one may determine the extent of decay of the ^{14}C and how long the water has been underground.

Vogel and Ehhalt[39] point out that ^{14}C is present in groundwater as dissolved bicarbonate, and, subject to certain conditions, residence times up to about 40,000 years may be measured. However, if true ages are to be determined, two basic requirements must be fulfilled: (1) the initial ^{14}C content must be known, and (2) the ^{14}C content must not change in the course of time by any means other than by radioactive decay. Before the ^{14}C content is used to measure residence time, it is necessary to determine whether or not the sample represents a "natural" case. The paper describes how the stable ^{13}C isotope provides much information on this question. Measurements of water from deeper levels indicate that low ^{14}C concentrations result primarily from decay and thus give true residence times. Analyses of water from oases and artesian wells in the Libyan Desert give ages mostly between 20,000 and 30,000 years. Figure II-6 shows these results. Two supplementary samples from shallow water galleries on the Mediterranean Coast are included for comparison.

In another paper Vogel et al.[41] describe and attempt to determine flow rates using the tritium content of underground waters of the Kalahari Desert. Since analyses of some samples did not show any significant quantities of tritium, the water must be considered to be older than 40 years. One sample contained a

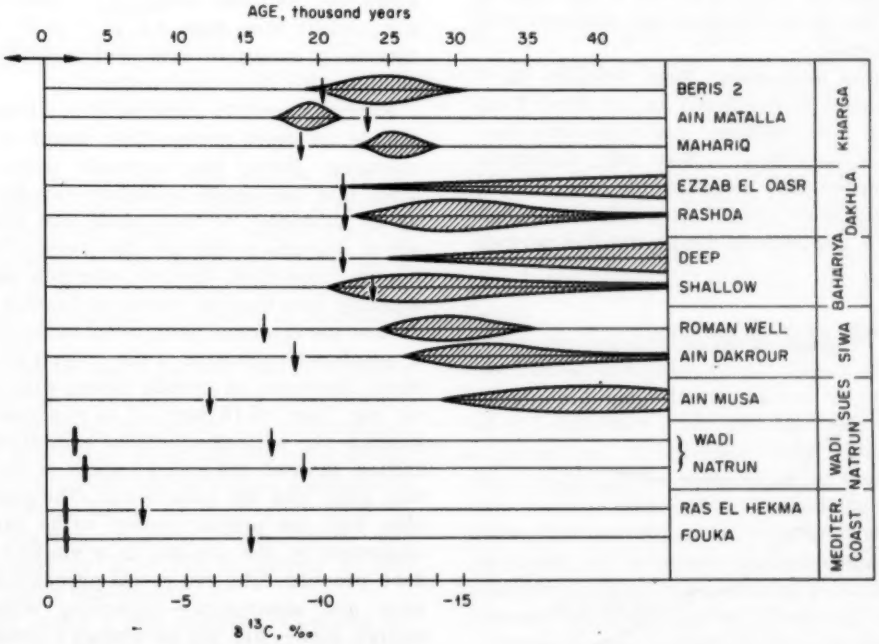


Fig. II-6 Carbon-14 ages of groundwater from Egypt (upper scale), and ^{13}C deviation (bottom scale, arrows).[39] The maximum error from the uncertainty of the initial ^{14}C content is denoted by two arrows on the time scale, and the probability distribution of the measured age is, with respect to counting error, represented by the hatched area. The ^{13}C scale represents the per-mille deviation, ^{13}C (‰), from the ratio of average marine limestone as a standard.

low but significant amount of tritium, indicating either an age of about 30 years or infiltration of a small amount of recent precipitation. From these results it was concluded that the recharge and flow rate for this catchment area are slow compared with the tritium half-life.

In a paper by Münnich and Roether (Ref. 40), ^{14}C and ^{3}H were used in a comparison of ages of groundwater. The purpose of the study was to determine whether or not there might be a decrease in ^{14}C content which exceeds that of decay alone and hence might be attributed to exchange.

MEASUREMENT OF RIVERBED VARIATION

In an application of isotopes that deviates from the usual tracer techniques in hydrology, Ariizumi and Kondo[31] used ^{60}Co and ^{137}Cs in backscattering densimeter probes to measure a boundary face between the water and riverbed layers. A tube was driven into the riverbed and supported at the top by a steel arm extending 5 m from the side of a bridge pier. As the densimeter probe was lowered within this tube, large variations were observed in the counting rate as the probe crossed the boundary face of the riverbed.

Using this instrument, these workers made measurements at Kurihashi on the River Tone in 1960, at both medium- and high-water conditions. During a flood in June 1961, variations in the riverbed were measured for several days. The results were plotted as a function of time (Fig. II-7), and several distinguishable periods were observed: that of scouring only, that of piling-up of a layer of low density accompanied by scouring, that of stable development of the piled-up layer, and that of contraction of the piled-up layer. These periods corresponded with a curve of the water level. A maximum scouring of 3.3 m with a high-water level of 5.5 m was observed.

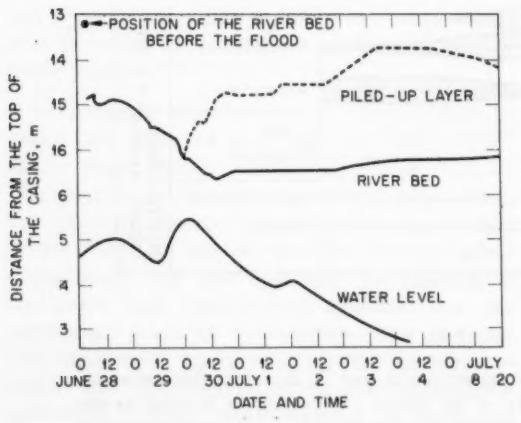


Fig. II-7 Relation between scouring and water level, June–July 1961 (Ref. 31).

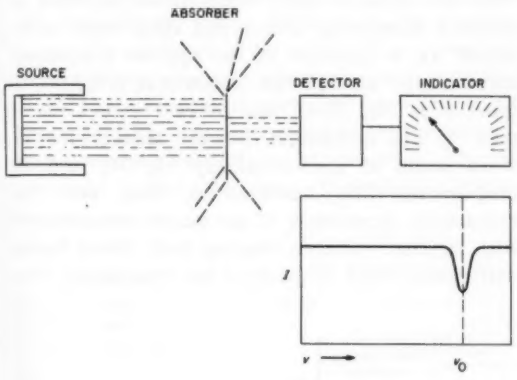
Practical Applications of the Mössbauer Effect*

Since 1958, when Mössbauer published his original paper[46] on an emission and absorption process for low-energy gamma rays, several hundred papers have appeared in the literature on this subject, and several conferences have been held. These low-energy gamma rays[47] are said to provide the most precisely defined electromagnetic frequency yet discovered.[48] The excited states of the so-called Mössbauer isotopes produce line widths of the order of 10^{-9} ev. When one compares this width (ΔE) with the energy (E) of the transition, e.g., ~0.01 Mev, then $\Delta E/E = 10^{-13}$. Thus an extremely sensitive (one part in 10^{13}) effect is available for many applications.

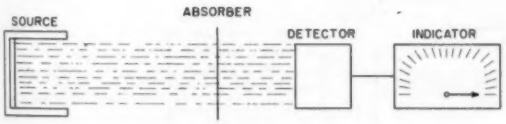
Energies usually considered in nuclear transitions are much greater than chemical bonding energies. Hence one normally thinks of the atom that undergoes a nuclear transformation as an unbonded, or free, atom. The Mössbauer effect depends on the fact that some of the low-energy transitions involve energies which are actually less than the chemical bonding energy, so that the chemical bond is not broken; instead, a quantized excitation of the crystal lattice occurs. However, in certain cases, with energies of less than ~0.15 Mev, it is possible to have gamma-ray emission *without* excitation of the lattice. Instead, the entire energy of the transition goes into the gamma ray. The gamma ray then has the proper energy to be resonantly absorbed by the crystal in a similar recoil-free process. The two recoil processes, emission and absorption—occurring without net energy loss—are the necessary processes for the Mössbauer effect. The normal response of a detector to the gamma radiation from a sta-

*J. J. Ezop, Illinois Institute of Technology Research Institute, reviewed the major portion of this manuscript.

tionary Mössbauer source, e.g., ^{57}Co, passing through an absorber, ^{57}Fe, is shown below.



When the source is mechanically moved or vibrated, a velocity, v, in the direction of the detector may effectively add the necessary energy for resonance absorption. The response is shown in the following diagram. Resonance ab-



sorption of the gamma rays occurs, and the de-excitation photons are emitted in a random fashion. Hence the count-rate meter shows a large reduction in the response. If one plots the intensity as a function of the velocity, a large drop in intensity is noted at resonance velocity (v_0).

Earlier work was directed toward a better theoretical understanding of the phenomenon. However, the great sensitivity of measurements using the Mössbauer effect and the wide variety of potential scientific and practical applications in such diverse areas as solid-state physics, nuclear physics, acoustics, mechanics, and space technology have led to efforts to find ways for using this effect.

Working under a DID contract, Ezop and coworkers[49] at Illinois Institute of Technology Research Institute (IITRI) undertook to study and evaluate the feasibility of using the Mössbauer resonance absorption phenomenon for solving practical problems in measurement and control technology. The use of such techniques for making small velocity, acceleration, and displacement measurements appears very promising. For example, velocities in the range 0.005 to 0.015 cm/sec are easily measured by ^{57}Fe nuclear resonance, and this range can be extended several orders of magnitude in both directions by using other sources and techniques. The IITRI workers used only ^{57}Co sources, with which the Mössbauer resonance in ^{57}Fe can be observed at room temperatures. However, the results of their investigations can be extrapolated to other Mössbauer isotopes, most of which require low-temperature apparatus to obtain an appreciable fraction of recoilless emissions and absorptions.

They showed that the direction and strengths of homogeneous magnetic fields of 300 to 1000 gauss may be determined by measuring the percentage of nuclear resonance in iron foils. With equipment arranged as shown in part *a* of Fig. II-8, they first measured the nuclear resonance and absorption as a function of the intensity of both the parallel and perpendicular polarizing fields between a source and an absorber. A 1-mc ^{57}Co source was placed in a magnetic field whose strength was varied between 0 and 1000 gauss, and the absorber, 0.7-mil-thick iron foil, was placed across the pole pieces of a permanent magnet with a field of 800 gauss. The rate of change of resonance absorption with magnetic field strength was greatest in the region between 300 and 1000 gauss (Fig. II-9). Nuclear resonance absorptions at magnetic field intensities of 1000 gauss were approximately 8.5% for perpendicular fields, 26% for parallel fields, and 15% for no fields.

The measurements were repeated with an electromagnet whose flux density was calibrated. A 2-mc ^{57}Co-Fe source was used, and the 0.7-mil-thick iron absorber was placed in a permanent 1950-gauss field. The results (Fig. II-10) were comparable to those in the first series. With parallel polarizing magnetic fields, the percentage of resonance absorption increased by a factor of approximately 2 at saturation, while perpendicular polarization decreased the percentage by a factor of 2.

Although the method is not, in general, as accurate as conventional techniques, such a system warrants further study for use when hostile environments or accessibility problems limit or prohibit the use of conventional techniques.

The results of preliminary studies showed that nuclear resonance techniques could be used to measure the amplitudes of acoustical (mechanical) motion in the megacycle frequency region. Also, indication was strong that such measurements could be extended to higher frequencies and possibly into the hypersonic (kilomegacycle) ranges where currently there are no suitable methods for measuring acoustical power or acoustical energy densities.

The experiments to determine the effect of high-frequency modulation of a Mössbauer source on the fraction of nuclear resonance absorption of gamma rays were made with a 2-mc ^{57}Co source plated on a 3-mil-thick No. 316 stainless steel foil and mounted on a quartz-crystal transducer. The intensity of the 14.4-kev gamma rays transmitted through a similar absorbing foil 1 mil thick was measured as a function of the applied frequency and voltage across the acoustical transducer (part *b* of Fig. II-8), and therefore as a function of the acoustical power. Measurements were made at approximately 10, 30, and 50 megacycles (the fundamental, 3rd, and 5th resonance harmonics of the quartz transducer) with applied voltages ranging from 0 to a value sufficiently high to destroy the resonance. The

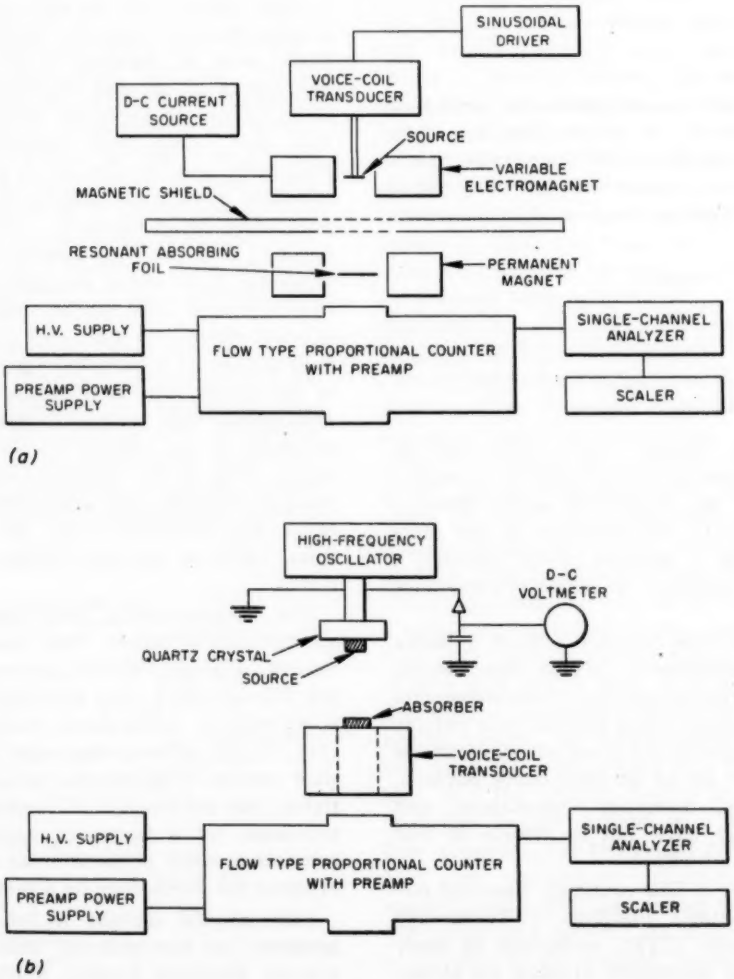


Fig. II-8 Experimental arrangement used to measure the effect of (a) *polarizing magnetic fields and* (b) *acoustical modulation on nuclear resonance absorption of gamma rays.*

relative intensities of these transmitted gamma radiations were then plotted as a function of the voltage at the transducer (Fig. II-11).

They further concluded that measurements of gravitational fields in interplanetary space or on the surfaces of celestial bodies, together with altitude measurements of space vehicles, by Mössbauer techniques are not practical. Measurement times required to obtain reasonably accurate results are of the order of hours or days under the most favorable circumstances. One possible improvement might be to use ^{67}Zn at cryogenic temperatures.

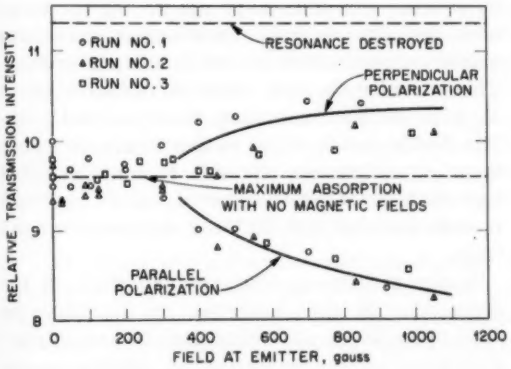


Fig. II-9 Transmission intensity of resonant gamma rays through an iron foil in a magnetic field of 800 gauss vs. magnetic flux density about a ^{57}Co source.

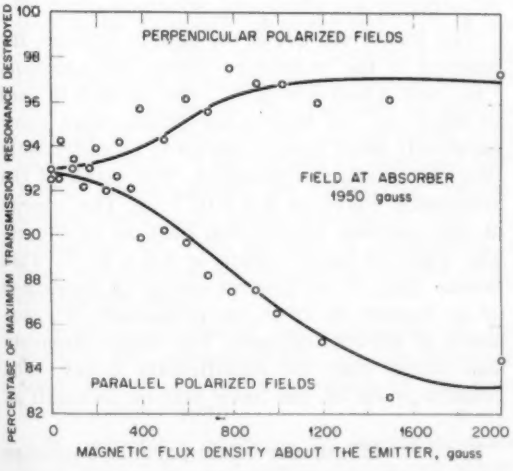

PERCENTAGE OF MAXIMUM TRANSMISSION RESONANCE DESTROYED
MAGNETIC FLUX DENSITY ABOUT THE EMITTER, gauss
PERPENDICULAR POLARIZED FIELDS
FIELD AT ABSORBER 1950 gauss
PARALLEL POLARIZED FIELDS

Fig. II-10 Percentage of maximum transmission of nuclear resonance gamma rays through an iron foil absorber vs. magnetic flux density about a ^{57}Co source.

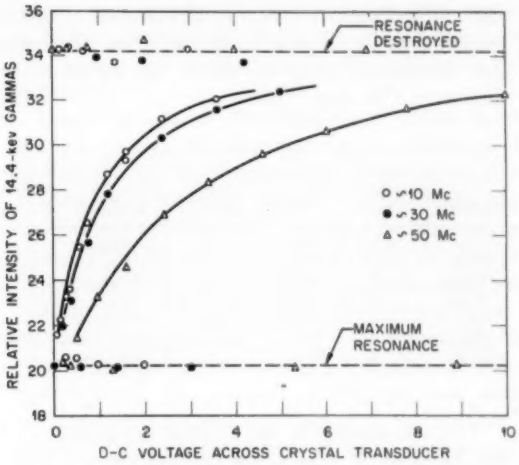


Fig. II-11 Relative transmission intensity of nuclear resonance gamma rays vs. applied voltage at emitter high-frequency transducer.

Likewise, pressure-change measurements by the Mössbauer technique used did not appear practical. No changes in resonance absorption were observed when a ^{57}Co–stainless steel source was subjected to pressures up to 40,000 psi. Theoretical calculations[50] indicate that pressures an order of magnitude greater than those used are required to observe a pressure effect in stainless steel.

Literature

The specialist in the Mössbauer field is familiar with the 1962 review by Frauenfelder.[51] More than 160 references are given, and numerous journal articles are reprinted for convenience. The extent of interest in the Mössbauer effect is indicated by the fact that the articles in this bibliography are in English, French, German, and Russian and come from half a dozen countries. Several review articles are readily available and can be read profitably by the nonspecialist.[52–55]

More recently the significance of the effect in chemistry and physics has been reviewed.[56–58] One of the more esoteric recent applications of the Mössbauer effect is for prolonging the half-lives of ^{119}Sn and ^{57}Fe (Ref. 59).

A bibliography compiled through Aug. 1, 1963, was published[60] in connection with the proceedings of the Third International Conference on

the Mössbauer effect. A breakdown of these references according to the number of papers on individual isotopes shows the greatest interest in ^{57}Fe and ^{119}Sn:

Isotope	No. of references	Isotope	No. of references	Isotope	No. of references
^{57}Fe	146	^{191}Ir	5	^{152}Sm	2
^{119}Sn	39	^{166}Er	4	^{170}Yb	2
^{197}Au	10	^{182}W	3	^{187}Re	2
^{169}Tm	8	^{183}W		^{193}Ir	2
^{61}Ni	7	^{151}Eu	2	^{155}Gd	1
^{161}Dy	7	^{129}I	2	^{159}Tb	1
^{67}Zn	5	^{149}Sm	2	^{83}Kr	1

For the novice in the field of applications of the Mössbauer effect, the translation of a Russian paper,[61] reprinted below, may serve as an introduction.

Applications of the Mössbauer Effect[61]

By B. N. Petrov, S. N. Bernov, and D. Ya. Libenson

One of the significant achievements of contemporary physics is the discovery in 1958 of the phenomenon of resonance absorption without recoil. This phenomenon is called the "Mössbauer effect" from the name of the German physicist Rudolph Mössbauer, who was honored for his discovery by the Nobel Prize in 1961.

The Mössbauer effect is a resonance phenomenon. Just as, for example, a string starts vibrating when a series of sounds is heard, the nucleus of a target can be excited by impingement on it of gamma quanta emitted by a nuclear source. A characteristic of nuclear resonance is its exceptionally great sharpness. However, nuclear resonance is not observed over a long period since the loss of energy in the nuclear emission disturbs the resonance conditions.

Mössbauer first discovered that, if the nucleus is not free but is bound into a crystalline lattice, there is a definite possibility for emission of gamma quanta without loss of energy in the emission. If, in turn, the target nucleus is also in a crystalline lattice, conditions are realized under which absorption of gamma quanta has a resonance character.

This can be shown in the following way. Let us assume that the emission is absorbed not by an individual nucleus but by the whole crystal. But the mass of the crystal is immeasurably larger than the mass of an individual nucleus; therefore energy loss in the emission becomes negligibly small. In the emission of photons, wave excitation sometimes arises in the crystal.

Naturally, if, in the excitation, gamma quanta of energy are transferred to the crystal lattice in waves, a corresponding digression from sharp resonance occurs. A change in the resonance curve may result from the effect of magnetic and electric fields on the atomic nucleus. The sharpness of the resonance in the Mössbauer effect is very great, and the magnetic and electric fields existing in solid bodies cause large changes in the resonance curves. This permits the use of the Mössbauer effect as a probe for examining magnetic and electric fields inside solid bodies in places where there is an atomic nucleus. It must be noted that such a "Mössbauer probe," if we may call it that, does not disturb the structure of a solid body.

X-ray structural analysis and study of the dispersion of cold neutrons are known to be very valuable in investigations of various phenomenons and the properties of different materials. Use of the Mössbauer effect makes it possible to obtain data that cannot be obtained by other means.

The special importance of processes of emission and absorption of gamma quanta without recoil is the extremely narrow resonance lines of emission and absorption or dispersion.

The longer the life of the atomic nucleus, the sharper is the resonance curve. For example, ^{57}Fe, with a half-life of 10^{-7} sec, is widely used in work on the Mössbauer effect. This comparatively short time is shown to be sufficiently long for a sharp resonance. The width of the resonance curve is 4.6×10^{-9} ev. The energy of the photons in a given case is 14,400 ev. The ratio of these values is 3.3×10^{-13}. This means that, if the gamma energy is decreased by an amount of 10^{-13}, the probability of resonance is greatly changed. For other elements this value may be significantly lower. The relative width of the lines may be as small as 10^{-18}.

Thus the Mössbauer effect permits a precise determination of the least change in energy, which was impossible a few years ago.

From this it is clear why, after the appearance of Mössbauer's first work, study of the

effect progressed rapidly, and at the present time there are several hundred journal publications on this subject. Studies in this field have been discussed at two international congresses.

Problems on the use of the Mössbauer effect in various fields of science and technology are now becoming important. These problems were recently discussed at a joint meeting of the departments of Technical, Chemical, and Physico-Mathematical Sciences of the USSR Academy of Sciences.

The use of the Mössbauer effect is dependent first of all on the interaction of a nucleus with the surrounding fields. Scalar Coulomb interaction leads to the so-called isomeric chemical displacement of the lines, and the electric quadripole and magnetic interaction results in hyperfine splitting of the line. Small velocities of the source relative to the absorber (Doppler effect), change in temperature, and gravitational interaction also result in line displacement.

The types of interaction mentioned make it possible to obtain information on the basic and excited states of the nuclei and especially on the medium surrounding the nucleus, which often is impossible by any other method.

The use of the Mössbauer effect for spectroscopic purposes is limited by the comparatively small number of nuclei that are suitable for studies by this method.

Spectrums of hyperfine structures make it possible to obtain the values of magnetic and quadripole moments of basic and excited nuclear states. Thus nuclear moments of ^{57}Fe and ^{119}Sn have been measured. Important evidence of changes in the radiuses of nuclear charges in excited nuclei can be found from measurements of the values of chemical isomeric displacements. By comparing experimental data with those calculated from the theory for the ^{197}Au nucleus, it is possible to draw conclusions concerning the nature of the nuclear potential.

From experimental measurements of the width of the resonance line, the life of the excited states can be determined (if the lines have a natural width).

An important problem for spectroscopists is searching for and putting to use new Mössbauer radiations.

Applications of the Mössbauer effect open up an extremely wide field in study of the properties of a solid body and in chemistry, since a larger number of different compounds and alloys may be studied with a single Mössbauer radiation.

Many aspects of the Mössbauer effect need further study. An important characteristic of the effect is the probability of emission (or absorption) of quanta without loss of energy in recoil. This probability depends on the spectrum of the frequency of vibration of the lattice of a solid body. In the case of a more complex lattice, the probability of the effect gives important evidence concerning the spectrum of possible crystal vibrations, i.e., the phonon spectrum of the crystal. Additional data may be obtained from measurements of temperature displacement of the lines. More complete information may be obtained in studies of the temperature dependence of the probability of the effect and the width of the lines and also anisotropy of the effect in monocrystals. Study of the effect of impurities introduced into a lattice of "foreign" atoms is very interesting.

From measurement of isomeric chemical displacements, evidence is obtained on the density of the electric charge in the region of the nucleus, and observation of the hyperfine structure of the absorption indicates the gradients of the electric field and values of the magnetic field acting on the atom.

The density of the electric charge on the nucleus and values of the electric field gradients are conditioned by the structure of the molecules and crystals, especially the chemical bonds. Study of the structure in ferromagnetic materials gives valuable evidence on the kinetics of chemical reactions, radiation changes in composition and properties of chemical compounds, the physical chemistry of polymers, and the mechanism of catalytic polymerization.

The Mössbauer effect also opens up new possibilities for radioactivation analysis.

Application of the Mössbauer effect to automation introduces new prospects for making highly efficient equipment for obtaining and processing information. In this field research is being promoted on development of the principle of construction of equipment for automatic control of small relative linear velocities and composition of complex mediums, which makes possible the measurement of velocities of the order of 10 μ/sec. Study of radiations with sharper resonance curves will permit, probably, making measurements of still slower velocities.

It must be noted that displacement of the resonance curve as a Doppler displacement makes possible the direct measurement of a velocity of motion, however small the value of the relative displacement. In this connection, use of the Mössbauer effect has known advantages over other methods, with which the velocity is shown only in the case in which the displacement approaches a definite value.

The next problem is to find appropriate Mössbauer elements with narrow lines for increasing the threshold of sensitivity and development of methods for increasing the rapid operation of the apparatus.

The use of the Mössbauer effect in automation, e.g., for control of pressure and temperature, is extremely promising. The possibility of using the hyperfine splitting of the Mössbauer lines in automatic-control equipment deserves more attention. Here, also, one of the important problems is the search for narrower lines and perfecting of methods of measurement.

A series of experiments on the Mössbauer effect is planned. In solid-state physics it will be used in studying, for example, types I and II phase transitions, the kinetics of phase transitions, and internal tensions and deformations; in chemistry, in the study of kinetics of chemical reactions, radiation-induced changes in the composition and properties of substances, paths of activation analysis, and peculiarities of chemical bonds and molecular structures; in automation and technical cybernetics, in automatic control of the chemical composition of complex mixtures and of small relative velocities, etc. This method is used also in geophysical and geochemical research and in prospecting for minerals.

Along these lines were directed, in a general meeting, the reports of V. I. Gol'danskii, "Prospects for Using the Mössbauer Effect in Chemistry"; P. L. Gruzin, "Use of the Effects of Gamma-ray Resonance Absorption in Metallurgy and Machine Construction"; D. M. Kagan, "Theory of the Mössbauer Effect and Questions of Solid State Physics"; V. S. Shpinel', "The Mössbauer Effect and Some of Its Applications"; N. N. Shumilovskii, "Prospects for Use of the Mössbauer Effect for Automatic Control"; and others.

The scale of the work on the Mössbauer effect and its applications, as was noted at the meeting, is still very small and for a long time will not meet the objectives of development work on the use of these effects in science and technology. A shortage of the necessary isotopes, pure monocrystals, and appropriate apparatus will create serious difficulties.

The problem of the scientific community and of the Presidium of the USSR Academy of Sciences is to assist in the wide development of work in the field of the Mössbauer effect and its applications and soon to call an All-Union scientific conference on this problem.

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Section

III

Isotopes and Radiation Technology

Process Radiation Development

The transmutation doping of semiconductors was first investigated in 1950 by Cleland et al.[1] using germanium. Since that time a considerable amount of work has been done in this area, including Lark-Horovitz's[2] study of the formation of lattice defects by nuclei recoiling from emission of decaying radiation, an investigation of annealing in germanium by Cleland and Crawford[3] and McKay et al.,[4] Fritzsche's study of impurity-banding phenomena,[5,6] and the preparation of uniform resistivity N type silicon by neutron transmutation by Tannebaum and Mills.[7] Crawford and Cleland[8] summarized in 1960 doping of various semiconductors and indicated the potential of this method.

The AEC's Division of Isotopes Development (DID) has supported work in this field for a number of years, and at least two AEC reports[9,10] and an abstract[11] have been published covering the efforts. An earlier issue of this review briefly mentioned[12] transmutation doping, and more recently an article by Klahr and Cohen[13] appeared in *Nucleonics* on the subject.

Klahr and his coworkers at Fundamental Methods Associates (FMA), Inc., have been engaged recently in research on fabrication of semiconductor devices by neutron transmutation doping. They have obtained precise spatial patterns of impurities deposited within semiconductor crystals. Some devices with rectifying characteristics have been obtained in a reproducible manner. Further development of this process could lead to the fabrication of complex devices and entire microcircuits at low cost by reactor irradiation of semiconductor crystals enclosed in a suitable "radiation die" of absorber material.

The following article reviews some of the principles of neutron transmutation doping, develops equations and discusses the economics of the process, and describes the FMA fabrication of operational electronic devices with this method. Klahr and Cohen's recent report[10] gives the detailed quantitative analysis for the characteristics of the devices produced.

Neutron Transmutation Doping*

Essentially the neutron transmutation doping process depends on two simple principles: first, the ability of thermal neutrons to transmute some semiconductor atoms into electronically active impurities, e.g., $^{30}Si(n,\gamma)\,^{31}Si \xrightarrow{\beta} {}^{31}P$; second, the ability of a thin sheet of cadmium with a slit pattern in it to absorb thermal neutrons, while permitting them to be differentially transmitted through the slits.

A silicon transistor consists of a silicon bar in which are deposited about one part per million of electronically active impurities in appropriate spatial patterns. These can be of either P type (positive current carriers, e.g., aluminum) or N type (negative current carriers, e.g., phosphorus). The simplest device is the diode, which has a single P-N junction with adjacent regions of different conductivity — P and N types. A transistor consists of two P-N junctions near each other.

In conventional techniques for making semiconductors, silicon crystals are uniformly doped with either P or N type impurities. Electronic devices are then made by inserting the opposite type of dopant in an appropriate spatial pattern by temperature-controlled diffusion of the dopant impurities. When complex devices are made in this way, the rejection rate is normally high.

*D. S. Ballantine, Brookhaven National Laboratory, compiled this article from reports of C. N. Klahr and associates of Fundamental Methods Associates, Inc., New York, N. Y.

The neutron transmutation process, on the other hand, can be a one-step irradiation procedure in which one can calculate the final electronic properties of the device to be fabricated. The process consists of the following steps:

1. Silicon that has been initially doped with P type impurities is placed in a special die.
2. The die, which is made of a material that strongly absorbs neutrons, is given a predetermined pattern of slits.
3. The die is placed in a holder, and the assembly is irradiated in a reactor whose characteristics are well known.
4. After irradiation, induced radioactivity is allowed to decay (usually a few weeks is required).
5. Neutron-produced defects and damage are annealed by heating.
6. The device is tested.

The results[10] demonstrate not only that the technique is practical but also that the calculated properties agree well with experimentally measured values.

The success of the transmutation doping process depends basically on the ability to produce a P-N junction. Factors affecting this ability are: (1) initial semiconductor properties, (2) reactor irradiation parameters, and (3) radiation die characteristics.

Figure III-1 is a simplified drawing of the essential features of the transmutation doping process and the irradiation device. Initially, there is a bar of silicon which has been uniformly doped with P type impurities. This bar is enclosed in a die of material that has a high neutron-absorption capability.

The die has a slit through which neutrons can readily enter in a straight line and react with the silicon to produce transmutations to N type impurities. The success of the process depends on achieving a final state in which there is a difference in electrical type between regions A and B. Obviously the device must be irradiated until region A has been converted to N type while region B still is P type (Eqs. 1 through 5, below). If we define the following:

N_0 = initial P type impurity concentration in silicon before irradiation

$N_{tr}(A)$ = added N type impurity concentration due to transmutation in region A beneath the slit

$N_{tr}(B)$ = added N type impurity concentration in region B under the die

$N_f(A)$ = final N type impurity concentration in silicon in region A

$N_f(B)$ = final P type impurity concentration in silicon in region B

it follows that

$$N_f(A) = N_{tr}(A) - N_0 \tag{1}$$

$$N_f(B) = N_0 - N_{tr}(B) \tag{2}$$

Equation 2 may also be written

$$N_f(B) = N_0 - (B/A)_G N_{tr}(A) \tag{2a}$$

where $(B/A)_G$ is a "geometric shielding effectiveness" of the radiation die, i.e., the ratio of $N_{tr}(B)/N_{tr}(A)$ of transmutations produced under the die to those produced under the slit. This value depends on the die thickness and slit geometry.

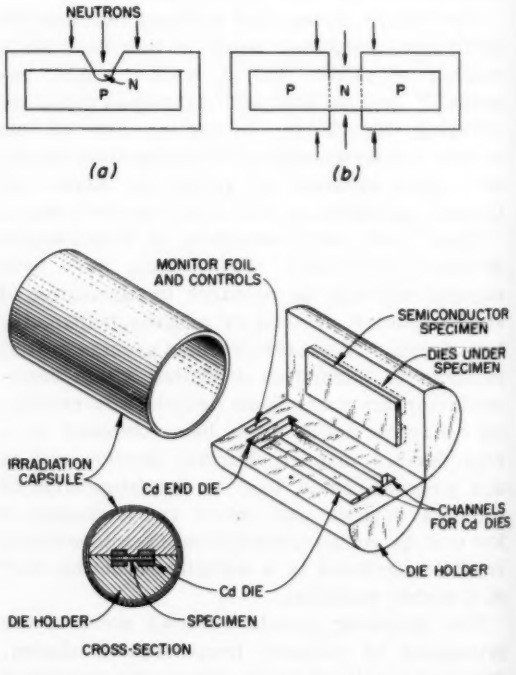

Fig. III-1 Schematic representation of neutron transmutation doping. (a) *Type A die.* (b) *Type B die.* (c) *Dies, specimen, and holder.*

The conditions for forming a P-N junction can be simply expressed as follows:

$$N_{tr}(A) > N_0 \tag{3}$$

and

$$N_0 > (B/A)_G N_{tr}(A) \tag{4}$$

or

$$(B/A)_G < 1 \tag{5}$$

Therefore, if $(B/A)_G$ is less than unity and Eq. 3 is satisfied, then the two requirements are met for production of a P-N junction by transmutation.

Formation of a P-N junction can be represented conveniently on a log-log diagram, "compensation doping diagram," as shown in Fig. III-2. Figure III-2 is based on an assumed $(B/A)_G$ value of 0.1 and an $N_{tr}(A)$ value of 10^{16} impurities/cm^3. From Fig. III-2 one can see that, for initial concentrations below 10^{15} impurities/cm^3, no junction is formed because both regions A and B become N type. Likewise, for initial concentrations above 10^{16} impurities/cm^3, both regions remain P type. In the initial concentration range, 10^{15} to 10^{16} P-N junctions are formed, and the final resistivities ratios can be read directly from the curves.

Another type of diagram which has been useful in predicting conditions under which P-N junctions will be formed is shown in Fig. III-3 in which $(B/A)_G$ is plotted against initial concentration, N_0. Since $(B/A)_G$ must be less than 1 and because $N_0 > (B/A)_G N_{tr}(A)$ is another necessary condition, it is apparent that the area between $N_0 = N_{tr}(A)$ and $N_0 = (B/A)_G N_{tr}(A)$ defines the range of values of N_0 and $(B/A)_G$ for which P-N junctions can be formed.

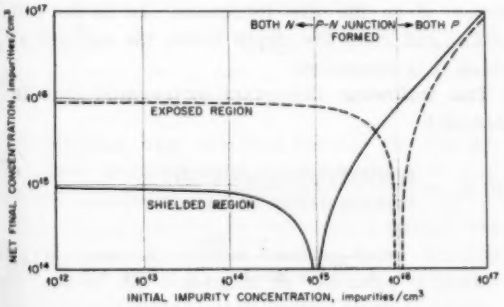

Fig. III-2 Compensation doping diagram.

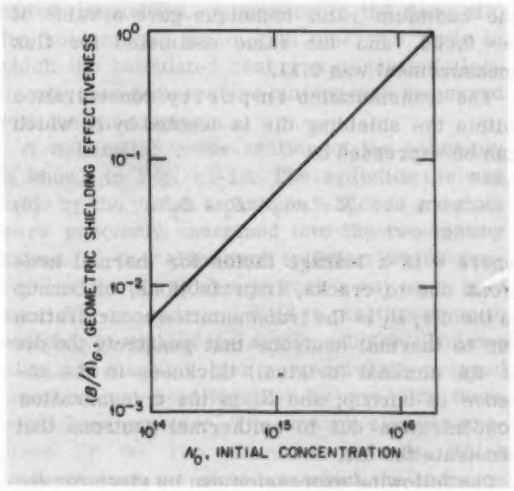

Fig. III-3 Relation between geometric shielding effectiveness and initial concentration of impurities. Any combination of N_0 and $(B/A)_G$ inside the triangle at top will produce a P-N junction. $(B/A)_G = N_0/N_{tr}(A)$; $N_{tr} = 2 \times 10^{16}$.

In planning experiments they considered (1) the uncertainty of the value for N_0, usually 5%; (2) the reactor flux uncertainty (limited to 5% for carefully mapped fluxes); and (3) the uncertainty in $(B/A)_G$, which is currently less than 25% and should decrease with experience.

Shielding effectiveness $(B/A)_G$ depends on die thickness, die material, and the reactor radiation spectrum.

The number of transmutations per unit volume per second in the unshielded semiconductor specimen is denoted by A. ($A = A_1 + A_2$, where A_1 transmutation concentration is produced by thermal neutrons and A_2 by epithermal neutrons.) It can be shown that $A_2/A_1 = 2\alpha$, where α is a parameter characteristic of the neutron-flux spectrum of the reactor. Approximate values of α can be calculated from values of the average thermal-neutron-absorption cross-section, the scattering cross-section, the thermal utilization, and the moderating ratio. It can also be determined experimentally by measuring the cadmium ratio of the flux or by using flux monitors and determining the ratio of fast- to thermal-neutron flux. For the Oak Ridge Research Reactor (ORR), which was used in the first transmutation doping experiments,

the cadmium ratio technique gave a value of $\alpha = 0.085$, and the value estimated for flux measurement was 0.11.

The transmutation impurity concentration within the shielding die is denoted by B, which can be expressed by

$$B = \gamma A_1 + B_1 + B_2 \tag{6}$$

where γ is a leakage factor for thermal neutrons due to cracks, imperfections, or burnup in the die; B_1 is the transmutation concentration due to thermal neutrons that penetrate the die at its nominal (original) thickness in the absence of burnup; and B_2 is the transmutation concentration due to epithermal neutrons that penetrate the die.

The following expression can be given for B_1:

$$B_1 = E_2\,(\sigma_m T)A_1 \tag{7}$$

where E_2 is a function corresponding to an isotropic distribution of neutrons incident on the radiation die and σ_m is the absorption cross-section of the die of thickness T. Thus $\sigma_m T$ is the die thickness in *absorption mean free path* at thermal energy. The die thickness is normally selected to make B_1 negligible, i.e., for $\sigma_m T = 4$, $B_1 = 3.2 \times 10^{-3} A_1$.

The function B_2 can be expressed conveniently as

$$B_2 = A_2\epsilon \tag{8}$$

where ϵ is the fraction of the epithermal flux that can penetrate the die and can be related to an effective cutoff energy E_c. The die acts as if all neutrons below E_c are absorbed, while all neutrons above E_c can penetrate without attenuation. Table III-1 gives typical values of ϵ and E_c for several thicknesses of cadmium.

As mentioned earlier, the value $(B/A)_G$ is instrumental in determining if a P-N junction will be formed. The geometrical shielding effectiveness depends on a geometry factor, G, which gives the ratio of the mean flux in the semiconductor under the slit to the unperturbed flux in the absence of the radiation die. This factor depends on slit width, radiation die thickness, configuration of the slit pattern, flux depression due to the die, and the angular distribution of the neutron flux at the surface of the die. Fxpressions have been worked out for the geometry factor for two types of die

Table III-1 EPITHERMAL TRANSMISSION OF CADMIUM PLATES*

Cadmium plate thickness, in.	Effective cutoff energy (E_c), ev	Fractional transmission (ϵ)†
0.005	0.25	0.316
0.010	0.35	0.266
0.015	0.40	0.25
0.020	0.44	0.238
0.030	0.50	0.223
0.040	0.55	0.213
0.050	0.58	0.208

*Taken from Dayton and Pettus.[14] An isotropic incident angular distribution has been assumed.

†The value of ϵ was calculated for thermal-neutron distribution at room temperature.

(see Fig. III-1*a* and III-1*b*): type A, which is designed to produce a transmutation impurity distribution parallel to a semiconductor surface and not extending through the specimen, and type B, which is designed to produce a transmutation impurity normal to the surface and extending through the specimen from one surface to the other. The principal physical difference between type A and type B dies lies in the die thickness and in the ratio of die thickness to semiconductor specimen thickness. A type A die will be thin in comparison to the specimen, e.g., 0.001 to 0.003 in. in comparison to 0.010 in., while a type B die will be of the same order as the semiconductor specimen, e.g., 0.010 to 0.020 in. compared to 0.010 in. The respective G values are

$$G_{\text{type A}} = \frac{\pi Z}{2a} \tag{9}$$

$$G_{\text{type B}} = \frac{\pi (H + Z)}{2a} \tag{10}$$

where H is the die thickness, $2a$ is the slit width, and Z is the depth below the surface at which G is evaluated.

The following important expression then is obtained:

$$(B/A)_G = \frac{\text{transmutation under die}}{\text{transmutation under slit}} = \frac{\gamma + 2\alpha\epsilon}{G^{-1}[1 + 2\alpha(1 - \epsilon)] + 2\alpha\epsilon + \gamma} \tag{11}$$

The expression can be measured directly, and it can also be calculated from reactor and

radiation die parameters. It determines the conditions under which a P-N junction will be formed.

Design and Fabrication of the Radiation Capsule

Tables III-2 and III-3 summarize the optimum physical and nuclear properties desired for the components for the transmutation doping process. For an initial experiment the selection of die material was quickly narrowed to boron or cadmium, both of which were used by Crawford and Cleland.[8] The rare-earth metals were eliminated because of their tendency to oxidize and give a loose oxide coating. Both cadmium and boron have attractive nuclear properties, and cadmium was finally selected because it is more easily machined.

Table III-2 PROPERTIES OF IDEAL RADIATION DIE

Physical	Nuclear
1. Ductile	1. Very large macroscopic thermal-neutron-absorption cross-section and $1/v$ epithermal absorption
2. Machineable	2. Mononuclidic
3. High melting point	3. Daughters should be stable and possess nuclear properties similar to parent
4. Able to be soldered	4. High density of absorbing centers per cubic centimeter
5. Good thermal conductivity	
6. Conducive to etch-forming	

Table III-3 PROPERTIES OF IDEAL DIE HOLDER

Physical	Nuclear
1. Ductile	1. Very low macroscopic cross section (both thermal and fast)
2. Machineable	2. Stable daughters or daughters that decay to stable form with short half-life
3. High melting point	3. Low specific gravity, i.e., low gamma-absorption coefficient
4. Able to be soldered	
5. Good thermal conductivity	
6. Nongalling	

Aluminum was selected for the die holder; its main disadvantages are its tendency to gall and its inability to be soldered.

The chief concern in the capsule design was the danger of excess heating from various processes of fast-neutron heating, gamma-absorption heating, and, in the case of boron, (n,α)-reaction heating. Cadmium melts at 310°C, and high heat can cause diffusion of the transmuted impurities. A mockup of the final die, die holder, and irradiation tube was made in which the calculated heat was generated electrically and temperature buildup was measured with thermocouples.

A schematic cross-section of the assembly is shown in Fig. III-1*c*. The radiation die was made by the "inlay technique." Niches and slots were precisely machined into the two mating halves of the aluminum cylinder, which constitutes the die holder, in such a way as to hold the 0.020-in. cadmium foils in position so as to completely enclose the semiconductor. The two halves of the die holder were carefully pressed together and held together by wrapping them with aluminum wire. The uniform slit was provided by the two aluminum islands, which project along the slots into which the cadmium was fitted. A thermal-neutron-flux monitor foil is placed in one slot, which runs outside the length of the die. This measurement permits calculation of flux depression factors.

The specimens used were 0.500 in. long, 0.150 in. wide, and 0.010 in. thick. This size was a compromise between capsule-size restriction and desirable size for resistivity measurements. The die holder was $1\frac{3}{4}$ in. overall, and the irradiation capsule was 1 in. in diameter.

Reactor Irradiation Requirements

The selection of a suitable reactor and irradiation dose was dictated by the following considerations:

The total neutron dose (nvt) needed to be high enough to produce sufficient transmutations in the semiconductor, yet not so high as to cause a high burnup of the radiation die. The neutron flux had to be high enough so that too long an exposure time was not required, yet not so high that a large amount of heat would be generated in the capsules and specimens. A high cadmium ratio was used to provide a well-defined flux pattern so that epithermal reactions were negligible. A reactor with enriched fuel elements was also desirable so that the flux depression in the neighborhood of the radiation die would be small. Finally, a reactor was needed which had experimental facilities that could (1) accommodate the capsule and (2) be utilized over controlled time intervals. Most of these requirements were met by the ORR where the irradiation experiments were performed.

Experimental Methods

Semiconductor specimens of silicon and germanium ranging in thickness from 0.008 to 0.020 in. and from 0.6 to 1.1 in. in diameter were procured. The single crystal wafers were cut into specimens of dimensions 0.150 ± 0.001 in. by 0.500 ± 0.001 in. by 0.0085 ± 0.0005 in., using a high-speed diamond saw.

The conductivity of type P on N was measured by a thermoelectric probe technique; resistivity was determined by a four-point probe method using a Dumas Instrument Company probe specially adapted to the present requirements, an Electronic Measurements Company constant voltage supply, and a K-3 potentiometer. The resistivity, ρ, is given by

$$\rho = \frac{V}{I} \times \text{form factor} \qquad (12)$$

Detailed evaluations of form factors for various specimen geometries have been given by Valdes,[15] Smits,[16] and others.

Once resistivity has been measured, the net impurity concentration (after compensation) can be calculated from the well-known expression, $N = 1/e\mu\rho$, where e is the electron charge and μ is the mobility.

Tables of $e\mu$ for silicon and germanium as a function of resistivity are available. The tabulations of Irvin[17] were used for silicon and of Cuttriss[18] for germanium.

Specimens were annealed in air prior to irradiation to determine the effect of heat-treatment on resistivity. No effect was observed for silicon annealed to 800°C or germanium at 300 to 400°C for 1 hr.

The specimens were loaded in various locations in the die and in the capsule. Appropriate flux monitors were also inserted in selected places to yield data on (1) *nvt* on the die surface, (2) total unshielded *nvt*, and (3) the cadmium ratio.

The capsules were irradiated for 3 weeks in the A-7 facility of the ORR. After irradiation the capsules were appreciably radioactive, and disassembly took place in a hot cell. The specimens were cool enough for measurements after 1 month, and the radiation level was down to background after 2 months. Only the cadmium showed appreciable postirradiation activity. The specimens at the time of testing contained considerably less than 1 mc of activity.

The specimens were first cleaned and etched. Conductivity type and resistivity were measured, and then samples were annealed (silicon to 800°C; germanium to 400°C). After the specimens were annealed, they were again cleaned, and the resistivity was measured. The cycle was repeated for various times and temperatures until a constant resistivity value was obtained.

Where specimens contained both P and N regions, resistivity of each region was carefully measured, and the P-N junctions were located by chemical staining. A two-point "probe-march" traverse of current vs. voltage characteristics was made along the length of the specimen. Oscilloscope display of this experiment revealed rectification characteristics whenever the probes were on opposite sides of the P-N junction. Figure III-4 shows the patterns obtained from a specimen having a P-N junction. Two probes about 0.04 in. apart make contact with the semiconductor surface to complete a circuit. The straight lines indicate that

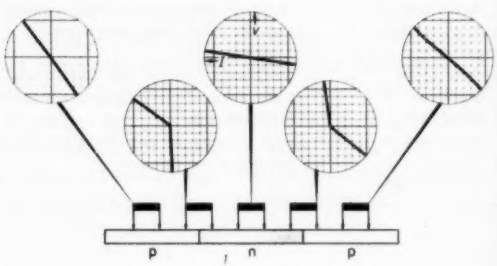

Fig. III-4 Sequence of oscilloscope displays in resistivity measurements of specimen having P-N junction. "Probe-march" method was used; voltage-current characteristics indicate where junctions occur.

both probes are in a P region and the "bent" patterns that one probe is in the P and the other in an N region. Note that (1) the resistivity is higher in the P region than in the N region and (2) the direction of rectification changes when the probe senses the change from P to N and then from N to P. Figure III-5 shows micrographs of two specimens.

Detailed measurements of junction characteristics were then made on properly prepared specimens. Voltage-current characteristics were measured using a K-3 potentiometer and a constant current source. Capacitance

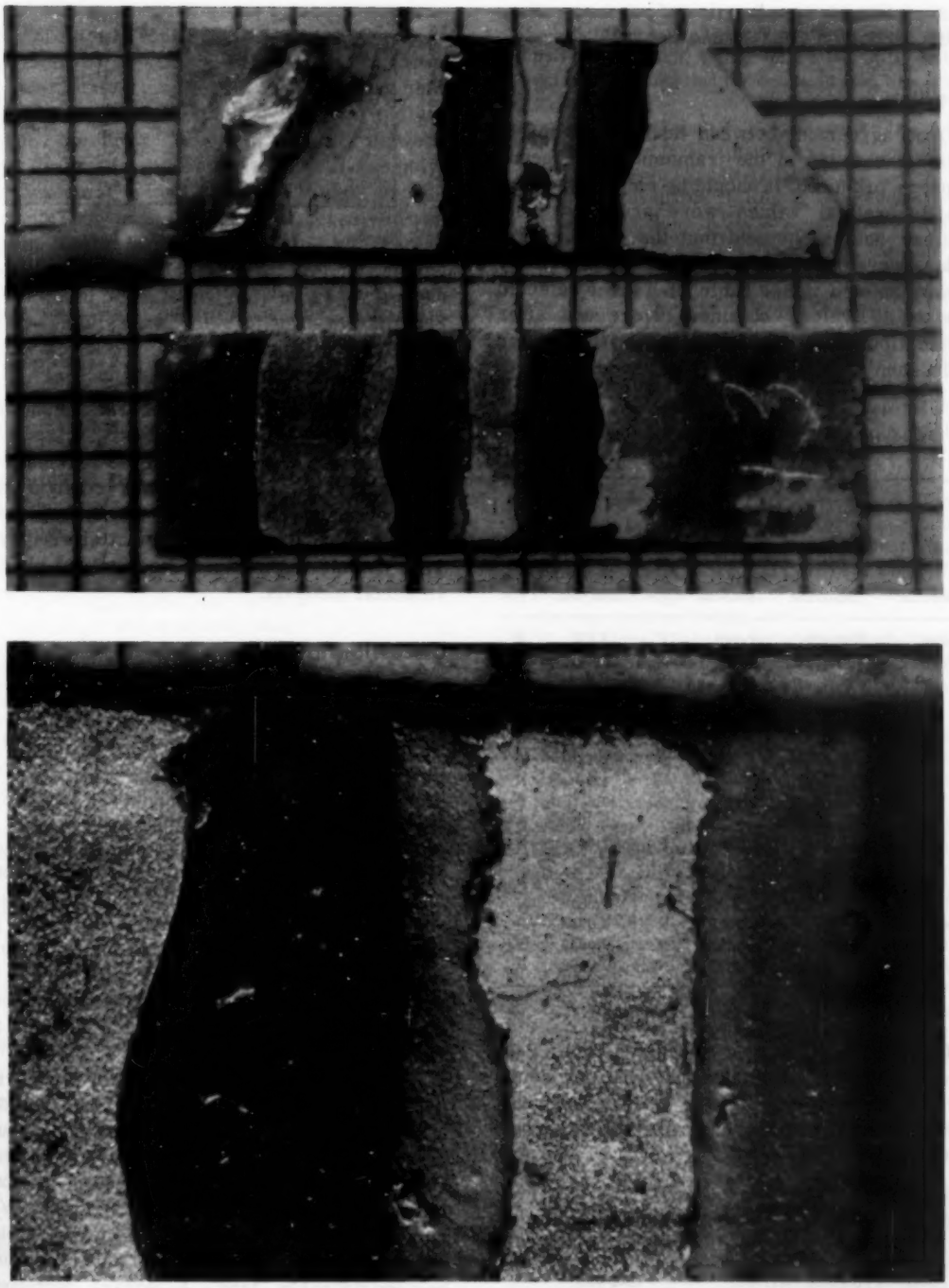

Fig. III-5 Micrographs of two specimens having P-N junctions.

was measured as a function of reverse voltage using a Boonton Capacitance Bridge.

Results

Good agreement between theory and experiment was found in the transmutation concentrations produced. It should be emphasized that the theoretical values were predicted from nuclear data alone and that the experimental values were obtained from electrical resistivity measurements alone. Table III-4 gives selected resistivity values obtained before and after irradiation. Detailed results are given in Klahr and Cohen's report[10] and are not repeated here.

Table III-4 RESISTIVITY BEFORE AND AFTER IRRADIATION

		Before irradiation		After irradiation	
Material	Type of exposure	Resistivity, ohm/cm	Type	Resistivity, ohm/cm	Type
Silicon	Uniform	0.405	P	1.07	N
		0.897	P	0.26	N
		1.48	P	0.214	N
		23.65	P	0.154	N
	Above die	20.1	P	0.252	N
		2.78	P	0.310	N
	Under die	0.799	P	2.14	P
	Under slit			1.18	N
	Under die	1.29	P	22.3	P
	Under slit			0.703	N
	Under die	2.505	P	1.04	N
	Under slit			0.51	N
	Under die	18.05	P	0.54	N
	Under slit			0.33	N
Germanium	Uniform	0.0058	N	0.0058	P
		0.124	N	0.0052	P
		0.00234	N	0.0102	P

The physical characteristics of the P-N junctions produced are also just what one would have expected. They compare well with conventionally produced junctions with typical properties given below:

Junction capacitance (zero bias):	17 pf
Junction width:	1.6×10^{-4} cm
Concentration gradient:	2×10^{18} cm^{-4}
Built-in potential barrier:	0.42 volt
Minority carrier lifetime:	>0.1 μsec

Capacitance with bias voltage follows the $\frac{1}{3}$ power of the voltage, just as one would expect for a linearly graded junction. This is shown in Fig. III-6.

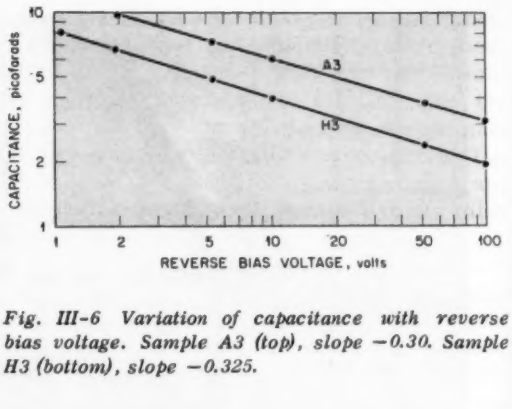

Fig. III-6 Variation of capacitance with reverse bias voltage. Sample A3 (top), slope −0.30. Sample H3 (bottom), slope −0.325.

An entirely novel feature of the P-N junctions produced was their geometric configuration. The junctions were normal to the specimen surface, as the radiation die pattern requires. This configuration cannot be obtained by conventional methods.

Discussion and Economics

The inlay method is probably only well-suited for one-of-a-kind experiments. However, a number of other methods have been considered including photoetching of radiation die configurations and casting of radiation dies (probably in vacuum) from a mold.

These approaches are being investigated now at FMA. Analysis of the irradiation economics indicates that microcircuits can be produced economically. A cost of about 30 cents per square centimeter of semiconductor is projected. A single reactor operating at 30 Mw could produce several hundred thousand microcircuits or other semiconductor devices per year.

A reactor in which 10 to 20% of the thermal neutrons are absorbed by "black" radiation dies could be of conventional size and not be much more expensive to build and operate than conventional research or test reactors. Although the upper limit on the irradiation cost for producing 1 cm^2 of microcircuitry in such a reactor is about 30 cents, the actual cost may be considerably less. A square centimeter corresponds to several circuits or devices. If the radiation die is made of natural cadmium, its material cost is nominal. If it is made of ^{113}Cd, the material cost for the die would

correspond to about 40 cents per square centimeter of microcircuitry.

A facility specially designed for neutron transmutation doping could be much like that of the ORR fuel assemblies. Ten fuel plates, each 0.1 in. thick with 0.1 in. of water between plates, would comprise a 2-in.-thick assembly. This assembly could have a square cross-section. Thus one would have successive rows of such fuel assemblies separated by irradiation cans. The difference between $n = 5$ and $n = 10$ would be reflected only in the size of the fuel assemblies, i.e., use of a 1- by 1-in. assembly for $n = 5$ or a 2- by 2-in. assembly for $n = 10$. In each case a radiation die plate would be placed between rows of fuel assemblies.

A total of 30 radiation dies, each with an area of about 70 by 70 cm = 5000 cm^2 could be irradiated in this reactor, which would operate at 30 Mw(t).

One cycle's throughput would be as follows:

$n = 10$: 30 radiation dies × 5000 cm^2 = 1.5×10^5 cm^2 of microcircuitry per cycle

$n = 5$: 3×10^5 cm^2 of microcircuitry

The cost of irradiation will be primarily dependent on the capital cost of the facility. An amortization cost of 20% of the capital cost per year and an operating cost, including hot-cell facilities, of an additional 20% per year were assumed. The total irradiation cost per year was estimated at $2 million for a $5 million facility, or about $40 thousand per cycle, for a 1-week cycle. This would correspond to a direct irradiation cost as follows:

$n = 10$: cost per $cm^2 = \frac{\$40 \text{ thousand}}{150{,}000 \text{ } cm^2} = 30$ cents per cm^2

$n = 5$: cost per $cm^2 = 15$ cents per cm^2

The cost per microcircuit would depend on the particular application, since the number of devices per square centimeter of semiconductor area would vary widely. A single circuit would probably not be larger than 1 cm^2, although it might be much smaller.

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Section

IV

Isotopes and Radiation Technology

Source Development

Large Sources of Beta Radiation for Chemical Processing

Current work in chemical processing with radiation from isotopes is mainly concerned with the use of gamma-radiation sources, notably ^{60}Co. Beta radiation, although studied for more than 12 years[1,2] for use in altering chemicals with a view to practical application, faces not only technical problems but also competition from alternate machine sources of electrons as well as from the efficient gamma-emitting isotopes. As matters now stand a number of gamma-radiation processing facilities have been constructed or are in progress—kilocurie research irradiators and others of commercial scale.[3] Beta irradiators akin to these units have been designed under a Division of Isotopes Development (DID) contract, but comparable ones are not being built. However, evaluation of beta-emitting sources continues, and indications are that they will find a niche in the rapidly growing field of radiation processing, especially for stimulating gas reactions and perhaps for treating surfaces or thin films. For example, in a current DID contract a reaction chamber and a 400-curie ^{90}Sr source are being used to evaluate hydrogenation by radiation catalysis. Recent attention in this work is focused on the commercially attractive process of telomerization of ethylene with methanol to yield higher molecular weight aliphatic alcohols (preferably C_{11} to C_{20}), which are useful as intermediates for biodegradable detergents.[4] Other research deals with the development of applications using ^{85}Kr as a radiation source and with basic reactions in radiation polymerization.[5]

The following paragraphs review the development and potential application of beta-ray-emitting isotopes for chemical processing, a study of vitreous enamels as source-bearers of three of these isotopes, and a number of experiments on grafting. Two types of beta irradiators are described, one in operation and the other proposed.

Development of Beta Irradiation

Like efforts on the possible industrial use of radiation in the chemical industry in general, work with beta irradiation has been concentrated in a relatively small number of fields. Although much work has been done in fundamental radiation chemistry,[6] many basic reactions that might be initiated by radiation have not been studied.

A recent reviewer[7] noted that a gap exists between research and application—between radiation chemistry and processing—and that a number of reactions have not been exploited:

> ... no mention has been made of the large number of studies carried out on the irradiation of hydrocarbons, alcohols, ketones and other classes of organic compounds which are largely of academic interest. It is, however, in the academic work on radiation chemistry that useful pointers can be obtained towards potential industrial utilization.

The potential uses of beta radiation necessarily parallel those of gamma and machine processing. For example, all the common monomers that can be polymerized by the use of conventional free radical catalysts can also be polymerized by radiation (see review by Roberts[8]); radiation-induced graft polymerization can be used to modify the surfaces of fibers for dyeing or to reduce their tendency to build up static charge; and radiative reaction

chambers can be used to induce halogenation, oxidation, sulfoxidation, cracking and alkylation of hydrocarbons, and addition reactions like production of ethyl bromide.[9]

Most of the effects in radiation processing are the same whether the electrons come from beta-emitting isotopes, electrical accelerating machines, or from gamma-ray reactions that produce high-energy electrons in passing through matter. However, penetration of gamma rays is much greater than that of electrons (Fig. IV-1; Table IV-1), and this characteristic, among others, dictates the choice between the two radiations. No beta-emitting radioisotopes, for example, qualify for radiation processing of packaged food. On the other hand, polyethylene sheet can be arranged to use up high-energy electrons from beta emitters or machines more efficiently than radiations from gamma emitters. Similarly, beta radiation can be used effectively to initiate reactions within vessels, especially when the radiation source is enclosed in the vessel.

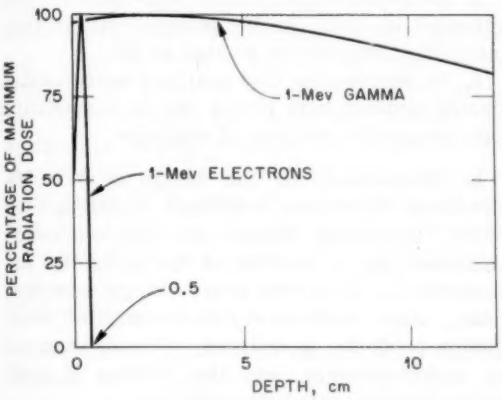

Fig. IV-1 Attenuation of 1-Mev gamma rays and 1-Mev electrons in water (from Jefferson[7]).

Table IV-1 ENERGY LEVELS OF DIFFERENT RADIATIONS*

Radiation	Energy, Mev
Visible light	10^{-6}
Ultraviolet light	10^{-6} to 10^{-5}
X rays	~0.01 to 5
Gamma rays	0.01 to 2.5
Beta particles (electrons)	10^{-5} to 250
Alpha particles	2 to 400

*From Hills.[9]

Rupp[10] has noted that the following factors generally influence the choice between machines and isotopes:

> Machines have the advantage of high intensity, on-off operations, lower initial capital cost, relatively uniform output and freedom from possible radioactive contamination. On the other hand, radioisotopes involve very little machinery, require no electric power, can be arranged in an infinite variety of geometric configurations, emit relatively pure radiation to produce large uniform radiation fields, and probably have lower operational and maintenance costs.

Morganstern,[11] on the other hand, pointed out that in the United States about 100 kw of electron-produced radiant energy is being used to every 0.5 kw in the form of ^{60}Co, the reasons being mainly the economical use of machines compared to isotope sources.

THE G VALUE AND CHEMICAL PROCESSING

The amount of material, A, obtained by a dose, R, of radiation can be related to the "G value" and molecular weight M by

$$A \propto G \cdot R \cdot M$$

in considering reactions that might be carried out economically.[7] The G value is defined as the number of molecules that react for each 100 ev absorbed in the system. Values of from 0.1 up to 10^7 have been obtained in various systems.

Emphasis in chemical processing has been on reactions with high G values or high molecular weight. For example, G values of about 10^5 are obtained in Dow Chemical's commercial production of ethyl bromide by addition of hydrogen bromide to ethylene, and extensive research on polymers has been done (see reviews by Bovey[12] and Charlesby[13] for estimated G values). In some of this work it was claimed that radiation was the only means of cross-linking;[14] however, irradiation products, produced abundantly, do not differ from those obtained by using cheaper sources of energy such as heat and light. Thus the search for new processes may turn toward developing nonchain systems, which, while giving lower overall yields, may give products different from those normally obtained.[9]

PHYSICAL AND CHEMICAL REQUIREMENTS OF LARGE BETA SOURCES

The properties required of large industrial beta-radiation sources have been reviewed in detail by Weinstock et al.[15] and those for smaller beta sources by the U. S. Department of Commerce:[16]

1. Long half-life isotope (1-year minimum), low gamma component, relatively high beta energies, and high yields (low attenuation)
2. Highly resistant to radiation damage
3. High specific activities (curies per unit area)
4. Low solubility in most normal solvents
5. High chemical stability
6. Good physical or mechanical stability
7. Capability of being manufactured by methods adaptable to remote-handling operations

SOURCE DEVELOPMENT

Only a few beta-emitting isotopes have been studied as potential sources in chemical processing, and some of these were specifically designed only for the study of radiation effects. For example, 14-hr ^{109}Pd was one of the early beta sources used by Brownell et al.[1] in a pressure reaction chamber or "bomb" at the University of Michigan to study the effects of radiation on chemical reactions. Later Meinke and coworkers[17] described the characteristics of 1200-curie sources made from ^{109}Pd foil. Brownell et al.,[2] in further work on the polymerization of styrene and natural oils, explored the possibility of using ^{90}Sr in various ceramic glazes. They noted that further work was needed on the glazes to develop them for high-temperature service, although they retained the radioactive ^{90}Sr at low temperatures. Seitzer and Tobolsky[18] also used ^{90}Sr as a point source to study the interaction of beta particles with organic liquids in the presence of vinyl monomers. Strontium-90 and a number of other beta-emitting sources are being used in radiation chemistry studies today. Volkova et al.,[19] in experiments on the irradiation of tetrafluorethylene in the liquid phase ($CHCl_3$), at 20 to 25°C with a ^{90}Sr source (dose rate, 5 rads/sec), found that there was a high tendency toward polymerization with a yield of approximately 10^6 molecules per 100 ev absorbed. And a 300-mc ^{85}Kr source in a glass bulb was used for several months in preliminary studies by Brown[20] on the irradiation of liquid materials.

Silverman,[21] reporting on DID-sponsored work at Radiation Applications, Inc. (RAI), at a 1959 IAEA conference on large radiation sources in industry, concluded:

1. There are promising areas for commercial application of fission-product beta emitters in the radiation processing field, particularly in the graft copolymerization modification of formed plastic surfaces and textiles.
2. Massive, rugged, inert, safe, inexpensive beta sources may be fabricated by suitable extensions of existing techniques. Source-bearing glass formulations show particular promise.
3. Beta-absorption calculations indicate that extended sources can be designed with power-utilization efficiencies as high as 20%.
4. An engineering cost analysis indicates that fission-product beta power can be competitive with alternative sources of radiation.

In preparation for the design of beta irradiators, Silverman developed equations (see below) describing dosage and beta-utilization efficiency as a function of the geometry and composition of various source-target systems. Later, other workers at RAI investigated three fission products as sources, vitreous enamels as source-bearers, and the process of graft polymerization with beta radiation.

Vitreous Enamels of ^{144}Ce, ^{147}Pm, and ^{90}Sr

The properties of the principal fission-product beta emitters and a discussion of their relative merits as sources was given by Silverman.[21] Weinstock and coworkers[15] selected ^{144}Ce, ^{147}Pm, and ^{90}Sr as the best ones, studied the physical and chemical properties of thin vitreous enamels containing oxides of these isotopes, and established source dosimetry patterns for ^{147}Pm and ^{144}Ce. A parallel study by these workers resulted in development of a radiation-induced graft polymerization process especially suited to the use of large beta sources for modifying the properties of polypropylene films and fabrics.

ENAMEL DEVELOPMENT

The Weinstock group prepared over 60 different enamel formulations. These were tested for coating (enameling) characteristics, resistance to attack by various solvents and aqueous solutions, and resistance to thermal and me-

Table IV-2 COMPOSITION OF BEST EXPERIMENTAL FRITS (WEIGHT PERCENT)*

Source	Sm_2O_3	CeO_2	SrO	SiO_2	Na_2O	ZnO	B_2O_3	K_2O	Cs_2O	BaO	Al_2O_3	Others
^{147}Pm-^{147}Sm	23.7	---	1.0	51.5	14.3	2.5	3.6	0.2	---	1.0	---	---
^{144}Ce-^{144}Pr	---	23.8	Same	Same	Same	Same	Same	Same	---	Same	---	---
	---	Same	Same	51.4	10.3	Same	Same	Same	4.0	Same	---	---
	---	24.0	Same	48	16.8	Same	4.2	Same	---	Same	1.0	---
^{90}Sr-^{90}Y	---	5.5	25	52.0	4.3	Same	Same	4.3	---	---	---	---
	---	5.3	24.3	50.4	4.2	2.4	4.1	4.2	---	---	---	2.9 Cr_2O_3

*Also included in frit were (wt.%): Li_2O, 1.0; CoO, 1.0; and Sb_2O_3, 0.2.

chanical shock. The best formulations contained 24 wt.% of the active component (SrO, CeO_2, or Pm_2O_3), 51% SiO_2, and varying amounts of nine other ingredients. The investigators concluded that some of these formulations were more resistant to chemical attack than Pyrex glass and could withstand considerable thermal and mechanical shocks (repeated quenches at temperature differences of over 600°F and repeated point impacts of over 10 in.-lb). Some of the best formulations and a few results from these tests are shown in Tables IV-2 through IV-4.

TEST SAMPLES

Most test samples were made by sintering the enamels on 20-gage (0.0375-in.), type 304 stainless steel. They were of various sizes: 1 by 1, 1 by 3, 3 by 3, 3 by 7, and 6 by 12 in. Some were either partly or completely coated with epoxy resin.

Large samples were made by imparting an appropriate curvature to a metal blank and then enameling it; such samples were flat longitudinally and retained only slight transverse curvature. The use of a $\frac{1}{8}$-in.-thick stainless steel base eliminated the curvature problem and resulted in samples with greatly increased resistance to mechanical shock.[21a]

Several other attempts by these workers proved unsuccessful or were abandoned after preliminary trials: the use of $SrTiO_3$, polyester resin, nepheline syenite, and Decalso as source enamels; the use of ceramic base materials, such as mullite and aluminum oxide; and the use of wire mesh as an enamel support. (Decalso sources have been discussed by Lewis,[22] and $SrTiO_3$, by Cochran, Bloom, and Schneider.[23] The emphasis, however, was on the development of power sources with these materials. The use of $SrTiO_3$ pellets in large beta sources is discussed later in this article.)

Table IV-3 PROPERTIES OF EXPERIMENTAL BETA-SOURCE ENAMELS[21a]

Material	Acid resistance,* % wt. loss	Mechanical impact resistance, in.-lb	Thermal shock resistance,† Δt, °F
Strontium enamels	3.0 to 4.1	5 to 9‡	6 to 700
Cerium enamels	2.4 to 3.1	7 to 10	6 to 700
Samarium enamels	3.6 to 3.9	-	6 to 700
Pyrex glass	4.1 to 4.5	-	-

*Two grams of –200 mesh powder exposed to 200 ml of 20% HCl for 24 hr.

†This property showed dependence on enamel-coating thickness, thicker coats having slightly lower resistance.

‡Values of 20 to 25 in.-lb were obtained with samples on $\frac{1}{8}$-in.-thick bases. All other impact values are for 0.0375-in. base material.

Table IV-4 SUMMARY OF EXPERIMENTAL CHEMICAL RESISTANCE DATA FOR ^{147}Pm ENAMEL*

Leach liquid	Temp., °C	Number and type of samples	Duration of test, days	Leaching rate, ppm/day	
				Range	Average
H_2O	Room	3 pellets	140	0.1 to 1.6	0.7
	Room	2 tablets	120	0 to 0.3	0.05
	65	4 pellets	60	3 to 30	10
	~97	4 pellets†	150	1 to 40	18
	~97	2 pellets‡	90	3 to 95	~70
0.01*N* HCl	Room	2 pellets	180	0 to 16	2.5
*p*H 4 buffer	Room	1 pellet	200	0.5 to 0.8	0.6
0.1*N* NaOH	Room	1 pellet	150	0.1 to 1.3	0.8
5% NaCl	Room	1 pellet	130	1.1 to 2.3	1.8

*Composition of enamel is shown in Table IV-2.
†Minus 200-mesh powder used for preparation.
‡Minus 100 +200-mesh powder used for preparation.

DOSIMETRY

Theoretical dose calculations of (1) the point source in a homogeneous medium, (2) the point source in a multiregion absorber, (3) a plane source, (4) an infinite slab source, and (5) an infinite line source were discussed earlier by Silverman.[21]

Patterns were obtained by Weinstock et al.[15] for enameled ^{144}Ce and ^{147}Pm sources containing about 0.1 mc/cm^2. Results indicated that these enamels would allow the fabrication of beta sources containing up to 5 curies of ^{147}Pm per square centimeter in a 4-mil-thick coating, up to 5 curies of ^{144}Ce per square centimeter in an 8-mil-thick coating, and up to 1 curie of ^{90}Sr per square centimeter in an 8-mil-thick coating. Such sources presumably could deliver surface and near-surface dose rates in the range of 1 to 10 Mrads/hr.

The radiation dose from a finite planar beta source in a uniform absorbing medium is obtained by taking a point source function and integrating it over a finite plane. The function used was a modified point source function of Loevinger,[24] which has been substantiated by measurements made by several observers with 12 different isotopes covering an energy range of 0.167 to 2.24 Mev.

A pseudo correction factor $f_{SA}(x)$, was obtained for self-absorption from the dose-distance curves of an infinite plane source of finite thickness and a thin plane:

$$f_{SA}(x) = \frac{D_p(x) \text{ of thin infinite plane}}{D_p(x) \text{ of infinite plane of finite thickness}}$$

where D_p is the dose, at point p, from a thin plane beta source and x is the mass thickness of absorber (grams per square centimeter) or distance from the source. The $f_{SA}(x)$ factors were calculated for selected distances. Assuming that for identical conditions of activity per unit area and enamel thickness these factors are valid for a finite slab source, the dose rate from a finite plane source of finite thickness, h, is

$$D_p(x,h) = \frac{D_p(x) \text{ of finite thin plane source}}{f_{SA}(x)}$$

Another method for obtaining a dose rate from a finite slab source can be secured by using self-absorption factors calculated as $f_{SA} = (1-e^{-\nu h})/\nu h$, where ν is the apparent absorption coefficient in square centimeters per gram and is given by a function obtained empirically from ionization chamber measurements on point sources in air. For 1-sq in. ^{147}Pm sources, dose-distance curves were calculated by both procedures and results showed good agreement between the two methods. Only the second method was used for ^{144}Ce sources.

Depth-dose characteristics of ^{144}Ce and ^{147}Pm were determined by film dosimetry, direct measurements with a Nuclear-Chicago Cutie Pie Model 2588, and an extrapolation ionization chamber. Figures IV-2 and IV-3 show comparisons of dose rates obtained from experimental sources and the theoretical calculations just discussed.

Strontium-90 rather than ^{144}Ce probably would be used in commercial application because of its pure beta emission, long half-life, and good beta energy levels. For this reason dosimetry data were translated into terms of equivalent ^{90}Sr source patterns (Fig. IV-4). Although the

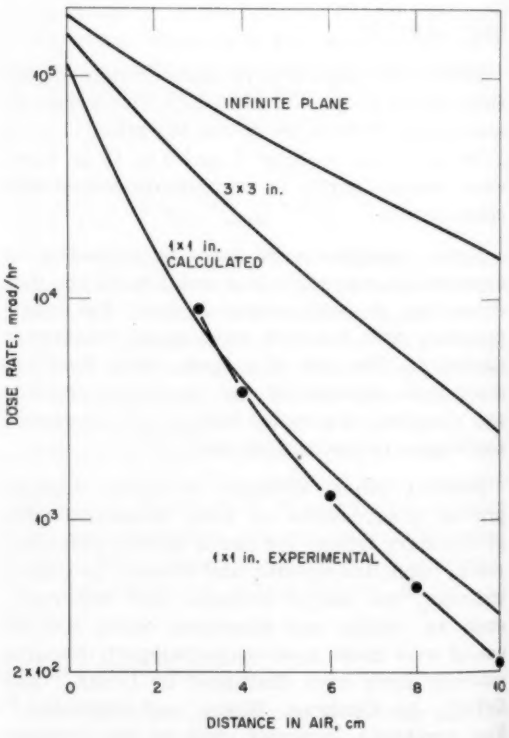

Fig. IV-2 Dose rate of 7.6-mil ^{147}Pm sources of various sizes. In these experimental data, 0.90 mc of ^{147}Pm per square inch was used.

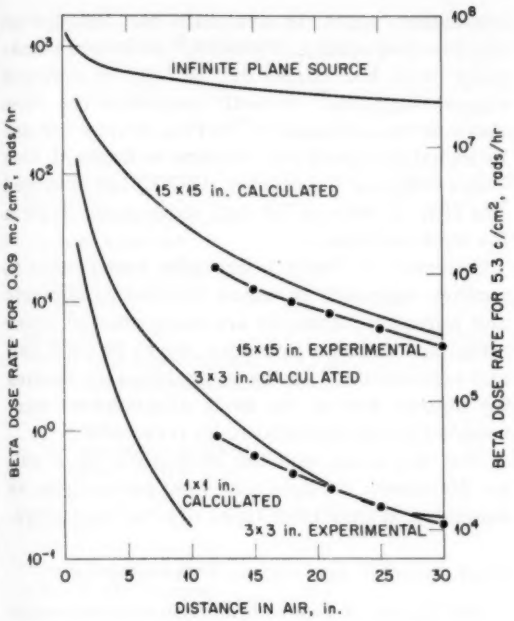
INFINITE PLANE SOURCE
15×15 in. CALCULATED
15×15 in. EXPERIMENTAL
3×3 in. CALCULATED
1×1 in. CALCULATED
3×3 in. EXPERIMENTAL
BETA DOSE RATE FOR 0.09 mc/cm², rads/hr
BETA DOSE RATE FOR 5.3 c/cm², rads/hr
DISTANCE IN AIR, in.

Fig. IV-3 Dose rate from ^{144}Ce sources.

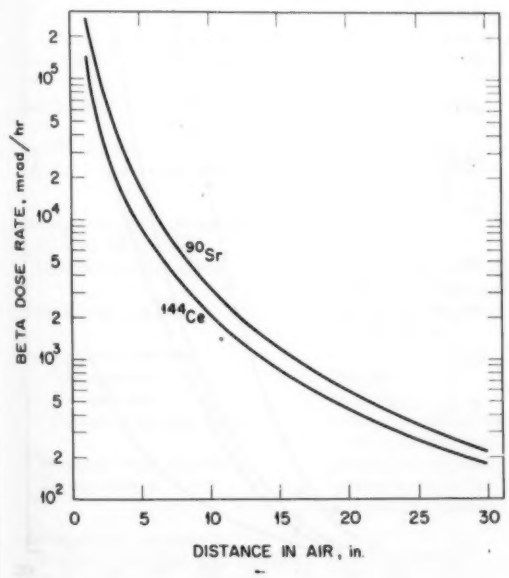

Fig. IV-4 Comparison of ^{144}Ce and ^{90}Sr dose rates. Doses were calculated on the basis of a 3-sq in. 8.44-mil-thick enamel (54.4 mg/cm²) source with a total activity of 5.1 mc. Self-absorption and backscatter corrections are included.

graph shows calculated depth-dose curves for both isotopes of identical source dimensions and activities of the two isotopes, it should be noted again that the beta-source enamels at maximum specific activity will contain 5 curies of ^{144}Ce per square centimeter but only 1 curie of ^{90}Sr per square centimeter.

Experimental Grafting Studies

Weinstock and coworkers[15] screened a number of systems for practical application, and a single one—a copolymer methacrylic acid (MAA)–styrene graft on polypropylene—was investigated in some detail.

Two methods were used in all tests: (1) mutual irradiation grafting and (2) postirradiation grafting. In the first, the polymer is irradiated while it is in contact with the monomer. In the second, the polymer is irradiated in air, vacuum, or nitrogen, and then immersed in the monomer.

Good grafts were obtained with a 2:1 MAA-styrene ratio. Also defined were the effects of a number of other process variables such as dose and temperature (Figs. IV-5 and IV-6) and pretreatments (Table IV-5). Dyeability and surface resistance of the resulting grafted samples seemed good. The dimensional change that occurs in MAA–styrene grafted films is important in designing a beta-irradiator pilot plant for graft copolymerization. The approximate increase in length and width of 2-mil polypropylene (grafted) was found to be as follows:

% graft	% increase
24.2	4.6 by 4.0
34.1	6.7 by 4.4
45.8	8.9 by 9.0

The methacrylic acid–styrene grafting system was applied to polypropylene fabric and dyed and undyed grafted samples shipped to four companies for evaluation of dry cleaning, washing, fadeometer, and other tests. Excellent dye retention and abrasion resistance was reported, but the fabrics showed very poor colorfastness. (Poor colorfastness was actually expected for the basic dyes used on the samples.)

Design of Beta Irradiators

Several beta irradiators have been designed, and a few research units are in operation.

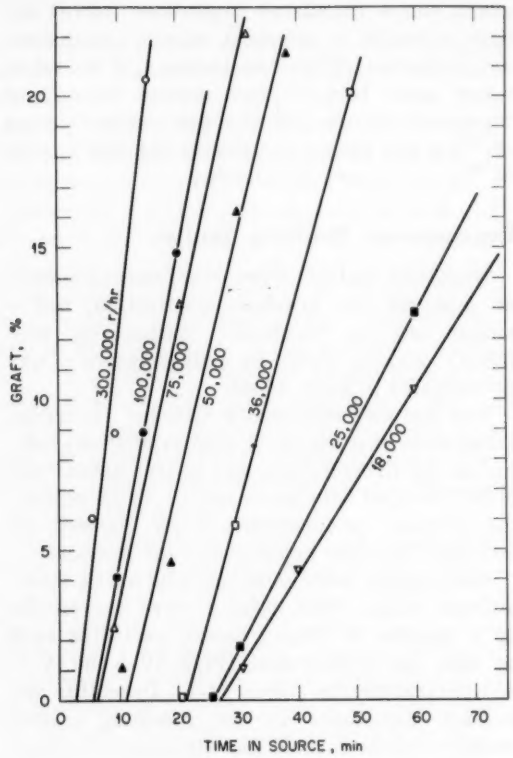

Fig. IV-5 Effect of dose rate on methacrylic acid–styrene (2:1) grafts on polypropylene.

Weinstock et al. at RAI proposed two—a commercial irradiator and a research irradiator.[25] Both would use enameled source elements. Yavorsky and Gorin,[26] whose work on the hydrogenation of coal has been reported in earlier issues of this review and elsewhere, for some time used 90-curie ^{90}Sr 3M Brand Radiating Microspheres[26a] inside a 10,000-psi, 500°C, heavy steel-walled reaction vessel. Recently they have installed a 400-curie $^{90}SrTiO_3$ source that provides about three times as much usable radiation power. Another advantage of $^{90}SrTiO_3$ is its ready availability. In the period 1961 to 1964, almost 5 million curies of ^{90}Sr was recovered from waste streams and converted to this form.[27] The ceramic has good thermal and radiation stability, reasonably high strontium content, and relatively low solubility in water.

A number of other beta irradiators have been built or are being considered. Johnston Laboratories BETALAB I, a megarad beta irradiator which is available for irradiation studies (especially of gases),[28] is loaded nominally with 600 curies of $^{90}SrTiO_3$ as sintered plates. Haybittle[29] recently published the dose rates from a 200-curie $^{90}SrTiO_3$ source for use in radiation chemistry studies in England. Oak Ridge National Laboratory (ORNL) has provided $^{90}SrTiO_3$ in both pellet and loose-powder form for such sources.

Sources of $^{90}SrTiO_3$ are also being used in another approach in which various monomeric and polymeric systems are being studied under simultaneous high pressure (up to 500,000 psi) and irradiation.[30] In these preliminary studies the source was in the form of a cylinder surrounded by the material to be irradiated.

The Yavorsky and the Weinstock units will be discussed in the next few paragraphs as examples of important types of beta irradiators.

FILM, FABRIC, AND PACKAGE IRRADIATORS

The design of research and commercial-scale beta irradiators was discussed in some detail by Weinstock et al.[25] Figure IV-7 shows the

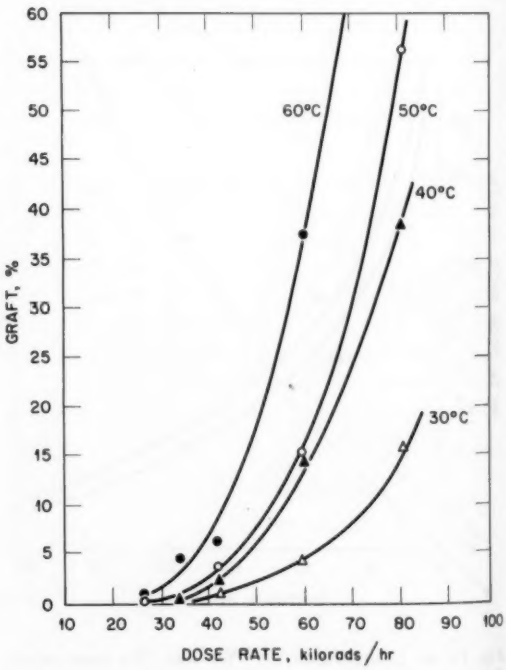

Fig. IV-6 Effect of temperature on methacrylic acid–styrene (2:1) grafts on polypropylene. Radiation time, 30 min.

Table IV-5 EFFECT OF FILM PRETREATMENT AND MONOMER-WETTED FILM IRRADIATION OF MAA-STYRENE GRAFTS

Film pretreatment	Graft, %	Dose rate, r/hr	Time irradiated, min
In styrene*		50,600	30
3-hr soak at room temp.	19.1		
60-sec dip at 80°C	23.6		
15-min dip at 80°C	27.4		
45-min dip at 80°C	23.3		
Control	18.3		
Other†		50,600	45
Film heated to 85°C, then immersed in comonomer	38.7		
15-min presoak in styrene at 80°C	50.7		
15-min presoak in MAA-styrene (2:1) at 80°C	48.7		
Control	34.0		
Wettings‡		97,000	45
23 at 2-min intervals	6.4		
12 at 4-min intervals	9.6		
8 at 6-min intervals	5.6		
	3.8		
6 at 8-min intervals	9.6		

*Percent graft as acid.
†Percent graft as sodium salt.
‡Species of graft not stated.

Table IV-6 DESIGN CRITERIONS FOR RESEARCH BETA IRRADIATOR

Isotope source	^{90}Sr or ^{144}Ce (5900 curies)
Source element	Multiple elements (5)
Source-element dimensions	13 by 6 in. (initial) 15 by 6 in. (future)
Arranged for film or fabric	
Film or fabric width	12 in.
Design irradiation rate	5 yd/min, but variable over a wide range
Monomer section	One or two monomer applications
Irradiation section	
Atmosphere	Nitrogen
Temperature	Ambient
Design residence time	3 to 4 min, but variable over a wide range
Monomer wash section	Two solvent washes
Drying section	Solvent evaporation (heated forced air)
Arranged for packages	
Maximum package size (approx.)	8 by 8 by $2\frac{3}{4}$ in.
Shielded entrance section	
Irradiation section	
Atmosphere	Air
Temperature	Ambient
Design residence time	1 to 2 min
Shielded exit section	

general arrangement of a research unit for film and fabric irradiation, and Fig. IV-8 is a sketch of the source-element cask and transfer cart.

There are four sections in the proposed unit: (1) monomer application, (2) irradiation, (3) excess monomer and homopolymer removal, and (4) drying. The material passes through the unit on a series of "live" or driving rolls with intermediate idler rolls for fabric support.

Incoming material passes through a section where monomer is applied by dipping in a tray. Additional monomer may be applied when the fabric is halfway through the irradiation period. In the irradiation section, fabric travels over a series of rolls past stationary source elements. The number of elements and design residence time of the fabric in the irradiator are based on an average integrated dose of 400,000 rads at the fabric design speed (see Table IV-6). Grafting occurs in the irradiation section in a nitrogen atmosphere at room temperature. A solvent wash removes excess or unpolymerized monomer or homopolymer. Residual solvent is evaporated in a warm-air drying section. When packages are irradiated, only the irradiation section is used.

A commercial beta irradiator similar in design to the research unit was also proposed by this group. Table IV-7 shows some of the estimated costs of this plant, which theoretically could process 15 million yd of material a year at about 3 cents/yd.

BETA IRRADIATION REACTOR

Yavorsky and Gorin of Consolidation Coal Co.[26] recently described in some detail the construction of a 90-curie beta irradiation reactor. Briefly, the source consists of 3M Brand Radiating Microspheres sealed inside 485 hollow stainless steel needles. The needles are cross-stacked in the reactor, and chemical reactants are circulated through the interstices by recycling. The recycling pump and reactor are shown in Fig. IV-9. The pump was devised so there would be no openings or mechanical connections, such as packing-gland shaft holes, to the outside. The piston is made of magnetic steel or iron and is actuated by a solenoid coil. The coil is wrapped around the nonmagnetic stainless steel pump cylinder. Piston-stroke frequency is controlled by a 200-volt electrical pulse from a motorized microswitch, in the

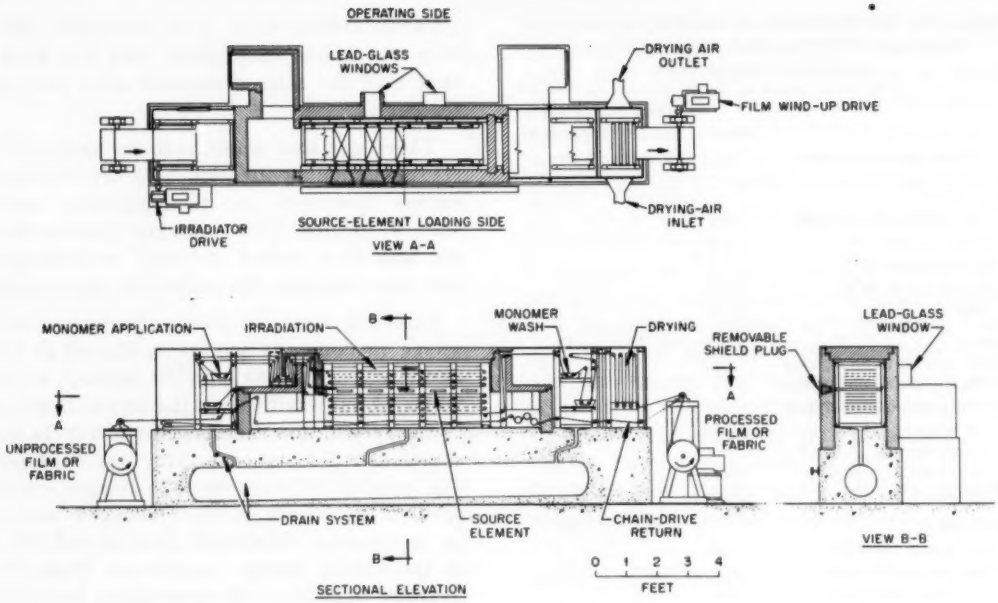

Fig. IV-7 Research beta irradiator proposed by Weinstock et al.[25]

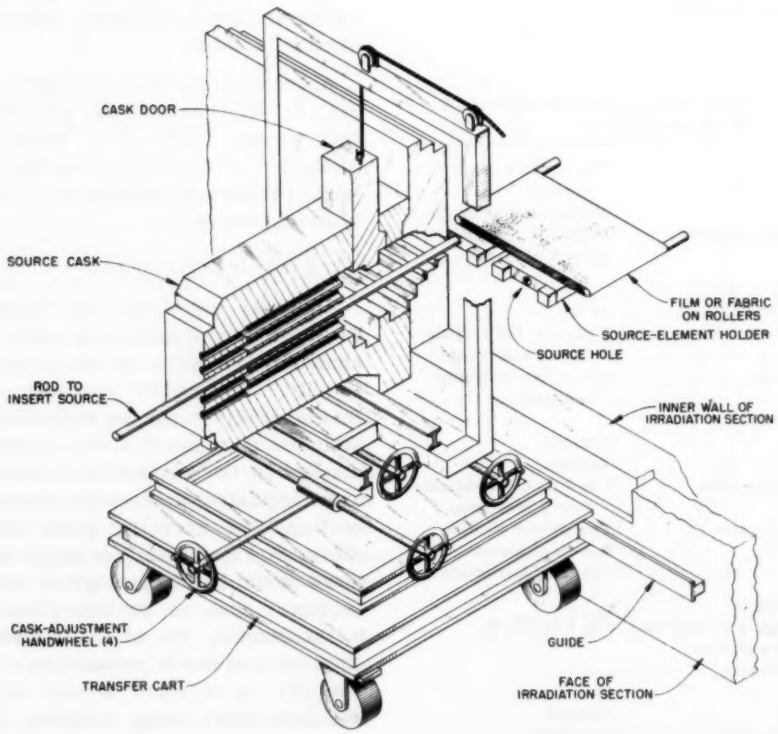

Fig. IV-8 Source-element cask and transfer cart in place at face of research beta irradiator.

range of 1 to 4 pulses/sec. Check-valve operation can be seen from the sketch. The upward or power stroke of the piston forces gas out of the cylinder, and gas refills the cylinder from the opposite end on the return stroke, which is driven by gravity or the piston could be spring-loaded for a horizontal pump. The feed enters the reactor at 160°C. It takes 132 min to reach a reaction temperature of 389°C and 155 min to reach 429°C.

Table IV-7 ESTIMATED INVESTMENT AND OPERATING COSTS OF COMMERCIAL BETA IRRADIATOR PLANT

Equipment (irradiator)	$265,200
Building	100,000
Engineering (10% of equipment and building)	36,500
Miscellaneous	10,000
Plant investment (*a*)	$411,700
Fixed operating costs	
Amortization (15% of *a*)	$ 61,700
Taxes and insurance (7% of *a*)	28,800
Interest (6% of *a*)	24,700
Profit (6% of *a*)	24,700
Total fixed operating costs	139,900
Variable operating costs (direct costs)	
Labor	
1 supervisor	$ 10,000
4 operators	48,000
1 mechanic	6,000
Variable overhead (50% of direct labor)	32,000
Material	
Methacrylic acid* at $0.40/lb	162,000
Styrene	30,000
Nitrogen	25,000
Miscellaneous	10,000
Total variable operating costs	$323,000
Total operating costs	$462,900
Cost per yard of material processed	
$\frac{\$462{,}900/\text{year}}{15{,}000{,}000\ \text{yd/year}} = \$0.031/\text{yd}$ or 3.1 cents/yd	

*Twenty percent of weight of film processed.

Fabrication of a swaged-tube 400-curie source recently was completed at Westinghouse (Waltz Mill). It consists of $^{90}SrTiO_3$ swage-compacted inside of 5-mil-wall stainless-steel tubing of 0.077 in. after swaging. The 9.2-ft-long tube is coiled in a helix, 1 by 4 in. The $^{90}SrTiO_3$ pellets were made at ORNL. The safety characteristics of this source have not been completely evaluated.

Since results with the 90-curie source were recently reported[26] and those with the larger source are preliminary, neither set is presented here.

Potential Future Applications of Large Beta Irradiators

Based on considerations of past research effort and proposed designs, large beta irradiators can be used effectively:

1. Where quality of radiation is a factor
   - *a.* In graft polymerization and modification of textiles and of plastic film or sheet
   - *b.* In vapor-phase synthesis of organic compounds in sealed vessels
2. Where beta irradiators are competitive with other electron sources and gamma irradiators because of such factors as
   - *a.* Economics—installation, operation, and maintenance
   - *b.* Portability
   - *c.* Amount of space available for shielding or storage of source
   - *d.* Heat production
   - *e.* Durability and safe operation

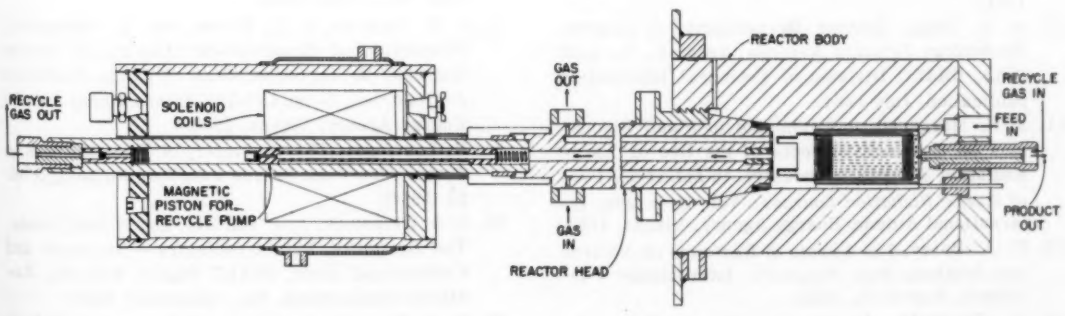

Fig. IV-9 Beta irradiation reactor with its enclosed recycling pump.[26]

It seems likely that significant developments in radiation chemistry may also direct the future application of beta irradiators. At present, cost factors and source form play a large role in their application to chemical processing.

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Section

V

Isotopes and Radiation Technology

Medicine

The Current Status of Nuclear Medicine in Japan

By Teruo Nagai, M. D.*

After interruption by the outbreak of World War II, radioisotopes began again to be imported into Japan in 1950, and the utilization of radioisotopes came again into the limelight after a 10-year interval. Since 1956, the field of nuclear medicine in Japan has developed at a logarithmic rate. For instance, the amount of imported ^{60}Co increased from 170 curies in 1952 to 189,000 curies in 1963 and that of ^{131}I rose from 1.26 curies in 1952 to 70 curies in 1963. Except in a few special cases, most of the imported radioisotopes were distributed through the Japan Radioisotope Association. About 80% of the total amount of imported radioisotopes is used for medical purposes.

On the domestic scene, some radioisotopes of shorter half-life, e.g., ^{42}K and ^{24}Na, have been produced in Japan Atomic Energy Research Institute's reactors and other reactors owned by private companies and by the reconstructed cyclotron. Recently, two pharmaceutical companies started producing various labeled compounds and radiopharmaceuticals commercially.

It is in biology and medicine that radioisotopes were first studied and are now used far more extensively than in any other field of utilization. Recent progress in the medical uses of radioisotopes has been greater than ever. Research and development of the medical application of radioisotopes are being carried on mainly at hospitals attached to universities, at national institutes, and at other hospitals.

Radioisotopes are used in routine techniques in basic research in biology and medicine. Particularly, ^{14}C- and ^{3}H-labeled compounds for various applications have come to be utilized quite extensively in recent years. This is also true in the field of clinical medicine, where the popularization of liquid scintillation counters and the uses of various ^{3}H-labeled compounds have attracted specialists' attention. The uses of ^{3}H-labeled compounds in microautoradiography have also developed rapidly. In particular, ^{3}H thymidine has come to be widely used in this field for studies of cell proliferation. Moreover, attempts are currently being made at electron-microscopic autoradiography. Compounds labeled with ^{3}H are also used in combination with thin-layer or gas chromatography in the study of biochemical reactions.

In the field of practical diagnosis, the most widely used kinetic study employing radioisotopes is the ^{131}I uptake test and scan for thyroid diseases. The T-3 resin uptake test has also become a routine technique in many hospitals. Other radioisotopes that have been widely used in examination of the functions of various organs include ^{59}Fe, ^{60}Co- or ^{57}Co-labeled vitamin B_{12}, ^{51}Cr, ^{131}I-labeled human serum albumin, ^{198}Au colloid, ^{131}I Rose Bengal and Hippuran, ^{203}Hg Neohydrin, ^{131}I triolein and oleic acid, and ^{131}I polyvinylpyrrolidone. New applications of one nuclide after another are being developed. Iodine-125-labeled Rose Bengal, Hippuran, human serum albumin, and erythropoetin, ^{132}I, ^{85}Sr, ^{132}Cs, ^{75}Se methionine, ^{57}Co EDTA, ^{131}I colloidal aggregates of albumin, radioactive ceramic microspheres, ^{131}I fibrinogen and fibrinogen antisera, ^{85}Kr gas, ^{15}O gas, and vari-

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ous ^{14}C- or ^{3}H-labeled steroids and other compounds have been put to practical use. Domestically produced radioisotopes of short half-life, such as ^{42}K and ^{24}Na, are now being used regularly for clinical purposes and whole-body counting. The application of neutron-activation analysis in the field of medicine is being studied also.

Measuring apparatuses, such as area scanners, linear scanners, medical spectrometers, and chromatogram scanners, have become as popular as X-ray diagnostic apparatus in routine use. Many manufacturers have developed equipment for practical use. Particularly noteworthy are the recently introduced color scanners and the magnetic-tape analyzers or memory scopes for analysis of "scintigrams." Positron scanners, scintillation cameras, human-arm counters, animal whole-body counters, low-background counters, and liquid scintillation counters have also been developed. Special devices, e.g., high-speed magnetic-tape recording systems, counters with built-in digital computers, and multichannel pulse-height analyzers, are being investigated to facilitate the study of hemodynamic processes. Human whole-body counters are utilized in ^{40}K measurements and in determination of tracer amounts for physiological and clinical diagnostic purposes.

The National Institute of Radiological Sciences, the National Cancer Center, and many hospitals attached to universities are now working on the development and application of radioisotopes to various diseases.

Reports and papers on research and development of radioisotopes are submitted to meetings and conventions of medical societies concerned and to the Japan Conference on Radioisotopes sponsored by the Japan Atomic Industrial Forum and to the annual meetings of the Japanese Society of Nuclear Medicine. The subjects discussed at the conference cover applications not only in medicine but also in all other fields. A number of European, American, and southeast Asian participants and observers have attended. The annual meeting of the Japanese Society of Nuclear Medicine has been held three times since 1961. At these meetings a number of subjects in clinical medicine are discussed. The number of papers presented at the meeting has been about 100 each year, and approximately 600 people hold membership in the Society. The meetings have been very valuable, and the proceedings of the meetings are published in English and in Japanese every year. The Sixth Japan Conference on Radioisotopes and the Fourth Annual Meeting of the Japanese Society of Nuclear Medicine were both held in November 1964 in Tokyo.

As for the education of physicians, since 1951 there have been six rounds of a 6-week course offered by the National Institute of Radiological Sciences with a total of 96 attendants. In addition, a similar course for pharmacists was started in 1964, and there will be an international course for the physicians supported by the International Atomic Energy Agency and the World Health Organization in the near future.

As outlined above, the use of radioisotopes in the field of medicine in Japan has made great progress, and today even small private hospitals can afford to establish diagnostic radioisotope laboratories. Moreover, some of the radioisotope techniques are now covered by health insurance. General practitioners and even pharmacists in hospitals have come to deal directly with radioisotopes. Many universities already have fully equipped laboratories of nuclear medicine, and it is currently planned to inaugurate courses in nuclear medicine for physicians and medical students at some large hospitals attached to universities.

Section

VI

Isotopes and Radiation Technology

Safety

Previous issues of this review have reported a Mexican radiation accident, the evaluation and testing of shipping containers and sources, and the shielding requirements for power sources. In addition, whole-body radiation exposures of personnel handling radioisotopes were listed in the first issue.

Over the years the nuclear industry has achieved an outstanding reputation for safe operation; radioisotopes are among the most safely handled of materials in the industry.

The first article in this section places in perspective the hazards of radioisotopes and other items in commerce. The second article presents the record of radioisotope accidents, and the third tells how the use of radioisotopes was "sold" to the public in one test.

Safety with Radioisotopes

By Sula J. Marsalis* and E. E. Beauchamp†

Despite the popular misconception of the inherent dangers of radioisotopes, these nuclides are actually less hazardous than many items of commerce and present less risk than many of the common situations encountered in everyday living. The 1920 disaster at Oppau, Germany, and the 1947 disaster at Texas City, Tex., killing more than a thousand persons and causing property damage of about $100 million, both involved a rather common fertilizer, ammonium nitrate. The fish kill and the chlorine spill on the Mississippi River were incidents that involved "business as usual."

Each year the accidental death rate is appalling; accidents rank fourth in the leading causes of death in the United States. In 1961 alone there were more than 92,000 accidental deaths, a rate of ~50 per 100,000 population. Of these, 37,000 were motor-vehicle traffic deaths (rate, 20.2), and 23,000 deaths (rate, 12.6) resulted from accidents in the home.[1] The ingestion of aspirin accounts for more deaths among children than any other single factor in the home.[2] Drownings accounted for 5000 deaths (rate, 0.7) in 1961, and in 1962 industry had nearly 14,000 fatalities and more than 83,000 permanent impairments from work injuries. From an overall viewpoint, in 1962 there were 27.9 accidents per 100 population. This number includes only the accidents that involved 1 or more days of restricted activity or medical attention, and of this number almost half (rate, 12.7) occurred in the home.[1]

Yet these incidents are accepted as part of normal living. On the other hand, loss of life and injuries from the handling of radioactivity have been negligible.[3,4] It is true that there were deaths and injuries resulting from the Hiroshima and Nagasaki bombs, and concepts about atomic energy in the public mind seem to be connected with these cities and with war. However, this loss of life was not caused by the handling of the radioactive materials.

Six plant workers have died from radiation injuries during the 20 years of the U. S. Atomic Energy Program.[5] (In that same period 1000 fatalities in this country have resulted from such things as bee stings and snake bites.) The AEC has had 35 lost-time injuries caused by radiation in these same 20 years. This was only 0.5% of the total AEC lost-time injuries. Considering the overall AEC safety record, an injury frequency of 1.92 per million man-hours contrasted with the national average of 5.99 per million man-hours, the injuries caused by radiation constitute an unusually low percentage.[6]

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The estimated 0.67-year shortening of the normal human life-span[7] as a result of working with radioactivity at the maximum levels permitted by law is far less than that from other causes. In Table VI-1 various factors that shorten the life-span are compared.[8,9]

Table VI-1 SHORTENING OF LIFE-SPAN BY VARIOUS FACTORS

Factor	Shortening, years
City vs. country dwelling	5.0
Single, widowed, or divorced vs. married status	5.0
Sedentary vs. physically active occupation	5.0
Overweight	
25%	3.6
35%	4.3
45%	6.6
55%	11.4
67%	15.1
Arteriosclerosis	
In 25th percentile of population having elevated lipoproteins	7.0
In 5th percentile of population having highest elevation of lipoproteins	15.0
Smoking cigarettes	
1 package per day	9.0
2 packages per day	18.0
All accidental deaths	2.3

Safety measures for protection of the workers and the public are taken in all stages of radioisotope handling—in production in a reactor or accelerator, in processing, in shipping, in a myriad of applications, and in final disposal as waste.

Special containment and ventilation, remote handling and shielding, and intensive operator training are provided in a radioisotope production and processing plant. When the material is readied to leave the plant to go to the user, each shipment is packaged and controlled in such a way as to protect from radiation every person who is involved in the handling and transportation.

Licenses are required for the shipment of radioisotopes, and specified government shipping regulations must be observed. The user—industrial firm, university, hospital, or laboratory—must likewise be licensed by the USAEC or by the state where this is now appropriate. General licenses are effective without the filing of applications with the Commission, but the specific licenses are granted only after proof has been given that definite requirements have been fulfilled. Waste disposal is also regulated, and maximum permissible concentrations released to the air, to sewerage, and to commercial disposal facilities are set by law.[10] The handling of radioactive materials therefore is regulated more carefully than that of many more familiar commodities.

The only completely unavoidable source of radiation is natural background, the most significant contributor to which is cosmic rays that originate in outer space. People are constantly exposed to these rays; at sea level and 50° latitude, for example, the dose rate to all body tissues is 50 mrem/year per person. The exposure nearly doubles for each 3000 ft in altitude (at ~7.5 miles, > 500 mrem/year).[6]

The second natural source of radiation is the radionuclides in the earth's crust, particularly ^{40}K, ^{14}C, ^{238}U, and ^{232}Th. Radiation from these elements totals an additional 50 mrem/year, with the actual dose also being affected by factors such as geographic location and type of housing. In some central areas of the United States, the radium content in bones is five times as great as in some other sections of the country; the terrestrial annual dose rate is 300 mrads in a granite house and 50 mrads in a frame house.[6]

Besides the cosmic rays and terrestrial radioactivity, there is the internal body burden of potassium, carbon, radium, and thorium resulting from the radioisotopes naturally present in the body and from inhalation and ingestion. This source contributes 30 mrem/year. Thus the total dose from natural radiation is ~130 mrem/year per person, and there is no escaping it.[6]

In addition to the natural background, there are many man-made sources of radiation other than the nuclear industry that increase the dose—some of these quite significantly. In the United States the average genetically significant dose from the mass chest X-ray program and diagnostic X-ray exposure is ~140 mrem/year and varies from 20 to 5000 mrem per individual skin dose depending upon equipment, techniques, and procedures used.[11] Mass chest X rays account for only 16% of the X-ray examinations in the United States, while 47% of the X-ray examinations are conducted by dentists. The total number of dental X-ray examinations of 0.4 per capita per year in this

country is considerably higher than that in other countries.[11]

Jet aircraft crews flying at 50,000 ft are subject to about 450 mrem/year; the crews of aircraft designed to fly at 85,000 ft (now on drawing boards) will receive 1500 mrem/year flying an average of 40 hr/month. Unnecessary exposure such as that with shoe-fitting machines, still not outlawed in almost half the states, gives doses as high as 7000 to 14,000 mrem per 20 sec of exposure.[12] Even commonly used items contribute to the accumulation. The average genetically significant dose to the wearer of a Swiss-made luminous-dial watch containing radium is ~8 mrem/year. The dose rate at the surface of a television screen is ~1 mrem/hr, but shielding reduces this to 0.5 mrem/hr at 5 cm from the screen.[6]

Thus the total average exposure of ~270 mrem/year from natural background and chest and diagnostic X rays alone is greater than the 233 mrem average work exposure at the Oak Ridge National Laboratory (ORNL), where a large number of the employees deal routinely with radioisotopes.[11] Since 1943 when ORNL operations began, only one employee has exceeded the permissible dosage determined* by the equation $D = 5(N-18)$ recommended by the International Commission on Radiological Protection (ICRP). This employee was involved in a radiation accident.[11] Exposures at ORNL should be typical of those in the nuclear industry.

Isotope Processing

The fact that work with radioisotopes is routine does not minimize the safety precautions required in their processing. Indeed, it is these safety precautions that prevent exposure, and certainly the best way to fight radiation injury is to prevent it. In all facets of radioisotope handling, exposure must be measured by secondary devices such as film badges, pocket dosimeters, pocket ionization chambers, or chemical dosimeters since the presence of radioactivity cannot be detected by the senses. Radiation can be detected by counters designed for particular types of radiation. Once a person knows the safety rules and accepts the reliability of the multiple secondary devices available for the detection and measurement of radioactivity, he no longer need be apprehensive about its use.

In the processing of radioisotopes, most problems of prevention relate to external exposure. Radiation exposures are ordinarily measured in units of roentgens (r) or milliroentgens (mr). One milliroentgen will produce an absorbed dose of ~1 mrad, and the biological effect of this absorbed dose of 1 mrad gives rise to the unit mrem.[13]

According to the National Committee on Radiation Protection (NCRP), the working limit of exposure of isotope handlers is 300 mrem/week.[14] The numerical values for exposure limits provide a conservative standard of safety, but lower exposure limits should be used if considered practical. The present rate for occupational exposure recommended by NCRP is 5 rem/year, which is equivalent to 0.1 rem/week for a 5-day workweek.[15] This rate is in agreement with the recommendation of the International Atomic Energy Agency (IAEA). For a person who is occupationally exposed at a constant rate from the age of 18 years, the formula $D = 5(N-18)$ implies a maximum weekly dose of 0.1 rem (or 100 mrem). It is recommended that this value be used for purposes of planning and design.[16]

Three fundamental factors can be used to limit exposure to external radiation: distance, time, and shielding. Distance is the most valuable since the radiation varies inversely with the square of the distance. Hence remote handling, that is, use of tongs and long-handled tools, and selective storing can make significant differences in radiation exposure. "Built-in" distance in packaging (Fig. VI-1), often referred to by the misnomer "space shielding," is used in shipping to cut down transportation costs of shielding equipment (50% of shipments from ORNL use this factor). However, safety can never be sacrificed for economy. Time is second in usefulness for protection from external exposure. Exposure is directly proportional to time. Careful planning prevents unnecessary lingering in the radiation field. If in certain types of production and processing the use of remote handling and the regulation of work time cannot be sufficient to ensure adequate protection, shielding must be used. All three of these protective measures are used according to need.

*D, tissue dose in roentgens equivalent man (rem); N, age in years.

Internal radiation is a second type of exposure that must be considered in safety precautions. Taken into the body by swallowing or inhalation, the isotopes, if they are absorbed

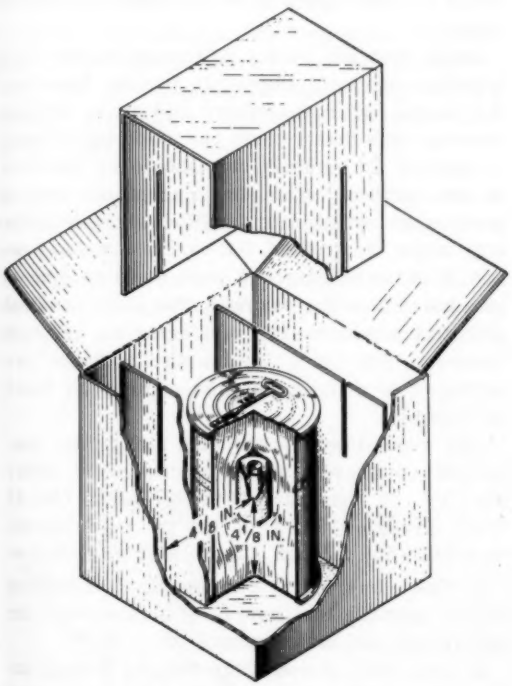

Fig. VI-1 Built-in distance in packaging.

through the walls of the stomach or intestines or lungs, spread to all parts of the body through the blood stream and irradiate the tissues involved. Ingestion is the most serious danger from radioactivity, since elimination of the radioisotope is sometimes difficult. Some isotopes are essentially completely excreted in days; others may be held for weeks or years, even far beyond the life-span.[17,18] Radioactive decay cannot be hastened, and the body's natural excretion can be hastened very little. Hence, preventing ingestion is one of the most important precautions to be taken by the worker. It should be mentioned that this difficulty of elimination also applies to certain household items such as caustic alkali, ammonia, and chlorine; the immediate and ultimate effects of these, too, can be very serious.

In order to prevent ingestion of radioisotopes, the worker must follow two simple rules: do not eat, drink, or smoke in a radiation area and do not use mouth-operated apparatus such as pipettes in any work with radioactivity. Supplementing this rule are the regulations designed to prevent inhalation, contamination, and direct exposure.

Tables of the more important isotopes have been provided by the AEC and NCRP for assessing any hazard.[17,19,20] Moreover, many publications are available in simplified and condensed form to enable easy reference to all aspects of safe handling of radioisotopes (see, for example, Refs. 20 and 21).

Processing of radioisotopes is performed in equipment designed to handle the maximum radioactivity involved. Reports dealing with construction of hot cells, hoods, off-gas systems, and waste-treatment equipment are readily obtainable.[22] Typical facilities are shown in Figs. VI-2 through VI-4. Small amounts of radioisotopes, particularly the alpha- and beta-emitting types, can be handled routinely with little shielding (for example, in glove boxes, Fig. VI-5). Gamma radiation may require very thick shielding in addition to remote handling to

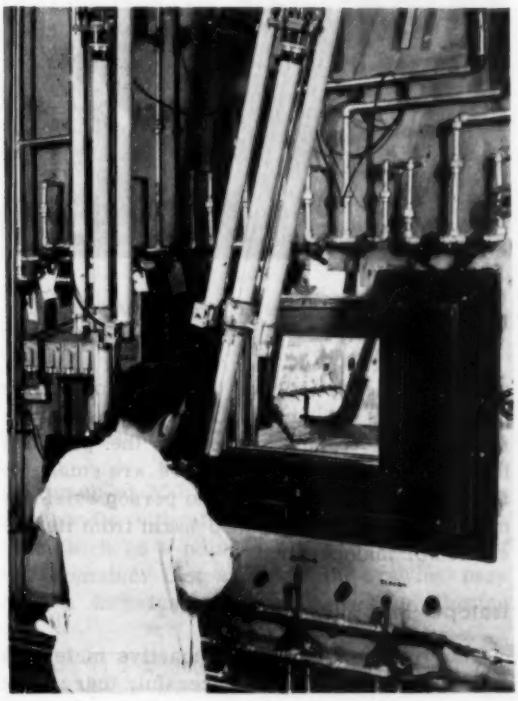

Fig. VI-2 Manipulator cell.

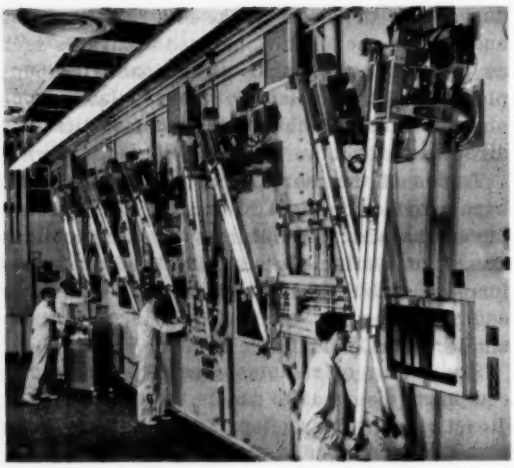

Fig. VI-3 Heavy-duty cells for development work.

Fig. VI-4 Loading cask in rear of shielded cell.

ensure protection from the radiation. Since processing is ordinarily carried out in adequate facilities by trained personnel, the potential hazards to the general populace are small. So far as can be determined, no person outside a nuclear plant has sustained harm from its operation or maloperation.[23]

Isotope Shipping

The transportation of radioactive materials to date has been quite successful; there have been very few accidents or incidents with any significant problems.[24] From ORNL alone, nearly 200,000 shipments containing a total of more than 2 Mc of activity have been made without significant incidents.[25] Add to these ORNL shipments the shipments from commercial suppliers, and the overall statistics would be many times more favorable than those cited.

More than 80% of the AEC radioisotope distribution-program shipments involve less than 2.7 curies of radioactivity and on an overall average less than 1 curie per shipment. If such a quantity of radioactivity should be involved in an incident, with few exceptions only a bothersome situation and public relations problem might be created, but hardly a major hazard. It is the shipment of large sources such as irradiated fuel, for example, that is the greatest potential hazard and should cause the greatest concern. For such shipments, routings are arranged to avoid population centers as much as possible.

The regulations for shipping radioactive materials commercially were prescribed under the Act of June 25, 1948 (62 Stat. 738; 18 U.S.C.A. 831-835). This law is implemented domestically through the following agencies:

1. The Interstate Commerce Commission (ICC), through the Bureau of Explosives, for rail freight and motor-vehicle service
2. The Civil Aeronautics Board, through the Federal Aviation Authority, for articles carried by air

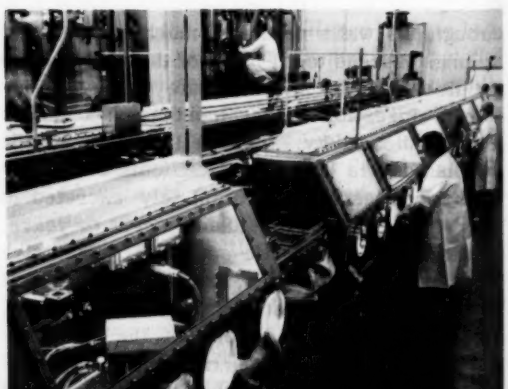

Fig. VI-5 Glove boxes for processing low-activity materials.

3. The U. S. Coast Guard for service by boat

When radioactive materials become involved in international operations, the following agencies provide appropriate regulations:

1. The International Air Transport Association (IATA) provides rules for air transport.
2. The International Atomic Energy Agency (IAEA) provides regulations for their own operations.

Several IAEA member countries have adopted these rules for regulation within their own countries.

Although for its own operations the USAEC is exempted by regulation from the ICC regulations, it nevertheless has adopted the policy of following these requirements. Futhermore, the Commission has had a profound influence on the requirements through its licensing operations and has promulgated broad regulations through the *Federal Register*. Particularly noteworthy now are AEC Manual Chap. 0527, which is proposed in the *Federal Register* as the well-known "Part 72," concerned with irradiated fuel elements. Likewise AEC Manual Chap. 0529, also proposed in the *Federal Register*, is concerned with "large sources." Figure VI-6 shows a typical shipping container used at ORNL. It illustrates the nonreturnable type, which accommodates approximately 80% of all radioisotope shipments from the Laboratory. Normally 2.7 curies is the maximum quantity allowed in such a container. This type of container evolved, beginning in 1950, to provide (1) a reasonable degree of safety from minor accidents during transport, (2) as little excess weight as possible to reduce shipping costs to the customer, and (3) a container that need not be returned, thereby eliminating the associated problems of inventory, check-in, maintenance, label removal, and any necessary decontamination. It is felt that the nonreturnable container has met these objectives through the tests of time and usage.

Figure VI-7*a* shows an ICC-55 type of container that is placed in a wooden box to simplify handling. This container is made with up to 3 in. of lead shielding and weighs a maximum of 350 lb, an amount that two people can handle without requiring a lifting mechanism such as a hoist. A further refinement of this container (not shown in the drawing) permits a 2R* insert to be used, thus allowing

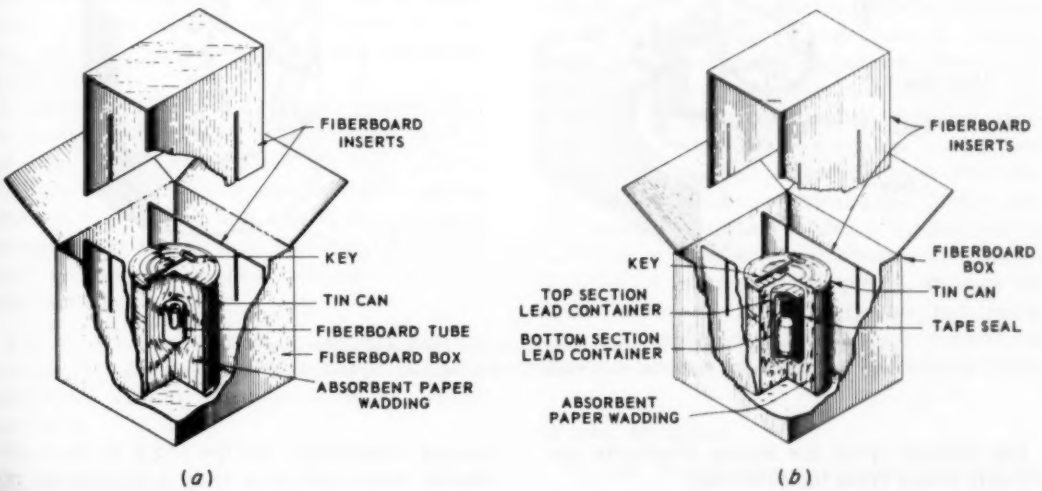

Fig. VI-6 Disposable containers for liquid shipments. (a) *Without lead shielding.* (b) *With* $\frac{1}{8}$ *to 1 in. of lead.*

*The 2R is a piece of stainless steel, malleable iron, or brass pipe made leakproof with screw caps at the ends and used to ship glass bottles containing liquid samples.

several radioisotopes to be accommodated in the same package.

Figure VI-7*b* is typical of a series of containers having from 4 to 9.75 in. of lead shielding and weighing from 300 to 6800 lb. These containers are used primarily for shipping ^{192}Ir, ^{60}Co, and ^{137}Cs.

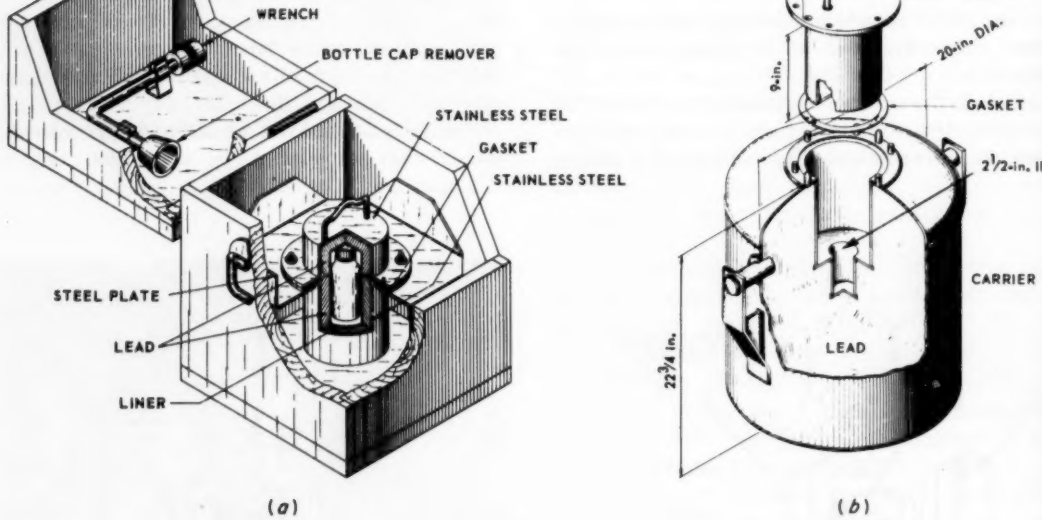

Fig. VI-7 Returnable shipping containers for (a) *liquid materials (lead shielding, 1 to 3 in.) and* (b) *solid materials (lead shielding, 1/2 in.), 2620 lb.*

The current trend for higher standards apparently stems from the following:

1. The realization that the future will require an increase in the shipments of irradiated fuel and large sources
2. A desire to ship irradiated fuel and large sources by conventional commercial methods without escort
3. Efforts on the part of the IAEA to prepare regulations that parallel those of the United States for the most part but in some cases are more restrictive than ours

The issuance of the proposed rule for Part 72 in the *Federal Register* (concerning regulations for containers to ship irradiated fuel) started a review of the problems involved in providing containers to withstand a maximum credible incident, yet built at reasonable cost. The shipping of irradiated fuel is a complex problem because of several factors: criticality, heat removal, and large quantities of radioactivity. The shipping of ordinary radioisotopes involves no problems of such magnitude.

There has been a desire to modify the ICC regulations to more nearly approach the regulations established by the IAEA so that ultimately there will be a truly international requirement. As a result of this, the Interagency Committee on Transport of Radioactive Materials has prepared some revisions and modifications which they have recommended to the ICC for adoption. At present this report is awaiting ICC action.

The IAEA continues to try to improve and to enlarge the scope of their regulations. An IAEA meeting, held in March 1963, gave consideration to adoption of a series of standard tests for containers. These tests were prepared by the United Kingdom. Some of them, such as the fire test, have merit; however, there is a need for design criteria and specifications for containers. Then a container could be designed and built with reasonable expectations that the container would meet the tests

that simulate transport conditions and, in fact, the test of actual usage.

The current major problem areas appear to be

1. *Fire Testing of Containers.* Both the United Kingdom and the United States are concerned with establishing a standard fire test.[26,27] The British approach appears to be somewhat less stringent, but it has the advantage of simplicity.[28] The British have introduced the concept of using hardwood as container-insulating material to withstand a fire. ORNL has performed tests on such shields. The work is summarized in an earlier issue of *Isotopes and Radiation Technology.* [27]

2. *Shock Resistance of Container.* Britain apparently has confined its efforts to a physical drop test. The United States is exploring prototype drop tests,[27,29] methods of calculation to determine acceptability, and the use of scale models for the physical tests in order to reduce the costs involved in producing full-scale prototypes.[30]

3. *Resistance to Piercing or Penetration.* The British have placed less emphasis on this test than the United States.[31] We have used a 15-*g* drop on a 6-in.-diameter object as the criterion.[30] However, this test appears rather stringent, and some other test may be devised which is probably more representative of an actual accident.

The concern and action of national and international agencies for consistent shipping regulations should further increase public confidence.

Isotope Applications

The widest applications of radioisotopes have been in industrial research and development, in plant and field problems, and in process or quality control.[20,32] The user of radioisotopes for any purpose must be licensed by the USAEC or, in some instances, by the state in which the work is done. The AEC issues both general and specific licenses.

The general license was granted to everyone when the regulations were first published. It permits the use of small, specified quantities of certain radioisotopes (adequate for most research projects) without special training, facilities for protection, or special application to the AEC. These "exempt" quantities of radioisotopes[33] are given in Table VI-2.

The specific license defines the isotope type, quantity, and limitations of usage. The requirements for a specific license are (1) an authorized usage, (2) availability of equipment and facilities adequate to protect health and minimize danger to life and property, and (3) availability of trained technicians with sufficient experience to qualify them to use the material requested.

The requirement of licensing is in itself the greatest safeguard against improper handling of radioisotopes. The Commission's regulatory program is directed toward public protection against the potential hazards from radiation by ensuring that the utilization, transportation, and disposition of radioactive materials are conducted in such a way that public health and safety are provided.[34,35]

Radioisotopes are cheap to use, and their applications are varied. Many new products and services have resulted from an initially small in-house isotopes program. New products and improved production methods, resulting from basic and applied research, usually mean repayment of initial cost many times.[36] But none of this would have been possible without safety considerations. Direct public benefits can accrue from applications of isotopes to industrial problems. The AEC's Division of Isotopes Development, through the ORNL Isotopes Information Center, has literature and references, available on request, on specific isotope applications in various fields.

Waste Disposal

By law, inventories of radioisotopes and records of waste disposal, shipments, receipts, and radiation exposure for workers must be kept. The regulations generally permit disposal of extremely low concentrations of certain radioisotopes directly into sewerage.[32] The maximum permissible concentration disposable by this means is set by law for each isotope; higher concentrations of waste can be handled through a waste-disposal service.

For many years land burial facilities at ORNL and at the National Reactor Testing Station in Idaho were available to licensees for the disposal of packaged low-level wastes. Several states now control radioactive-waste burial grounds.[37] Commercial firms licensed to receive and dispose of radioactive wastes are listed in Table VI-3.

Table VI-2 EXEMPT QUANTITIES OF RADIOISOTOPES

By-product material	Not as a sealed source, μc	As a sealed source, μc	By-product material	Not as a sealed source, μc	As a sealed source, μc
Antimony-124	1	10	Palladium-103–rhodium-103	50	50
Arsenic-76–arsenic-77	10	10	Phosphorus-32	10	10
Barium-140–lanthanum-140	1	10	Polonium-210	0.1	1
Beryllium-7	50	50	Potassium-42	10	10
Cadmium-109–silver-109	10	10	Praseodymium-143	10	10
Calcium-45	10	10	Promethium-147	10	10
Carbon-14	50	50	Rhenium-186	10	10
Cerium-144–praseodymium-144	1	10	Rhodium-105	10	10
Cesium-137–barium-137	1	10	Rubidium-86	10	10
Chlorine-36	1	10	Ruthenium-106–rhodium-106	1	10
Chromium-51	50	50	Samarium-153	10	10
Cobalt-60	1	10	Scandium-46	1	10
Copper-64	50	50	Silver-105	1	10
Europium-154	1	10	Silver-111	10	10
Fluorine-18	50	50	Sodium-22	10	10
Gallium-72	10	10	Sodium-24	10	10
Germanium-71	50	50	Strontium-89	1	10
Gold-198	10	10	Strontium-90–yttrium-90	0.1	1
Gold-199	10	10	Sulfur-35	50	50
Hydrogen-3 (tritium)	250	250	Tantalum-182	10	10
Indium-114	1	10	Technetium-96	1	10
Iodine-131	10	10	Technetium-99	1	10
Iridium-192	10	10	Tellurium-127	10	10
Iron-65	50	50	Tellurium-129	1	10
Iron-59	1	10	Thallium-204	50	50
Lanthanum-140	10	10	Tin-113	10	10
Manganese-52	1	10	Tungsten-185	10	10
Manganese-56	50	50	Vanadium-48	1	10
Molybdenum-99	10	10	Yttrium-90	1	10
Nickel-59	1	10	Yttrium-91	1	10
Nickel-63	1	10	Zinc-65	10	10
Niobium-95	10	10	Beta and/or gamma-emitting by-product material not listed above	1	10
Palladium-109	10	10			

Disposal of even high-level wastes does not create a problem at present. For 18 years great quantities of high-level radioactive wastes have been stored in tanks at Hanford and at Savannah River. The high-activity waste-tank farms are under constant surveillance; and, while a total of nine tanks out of 182 have been withdrawn from further service or given limited use because of possibility of leaks, there never has been any significant contamination of the earth surrounding the tanks.[37] Disposal of low-level wastes can be handled with little concern simply by following specified safety regulations.

Conclusions

It is axiomatic that no person wishes to be exposed to excessive doses of radiation. No person wants to subject himself to somatic injuries from which he may suffer; nor does he want his offspring to suffer from genetic injuries—injuries that may manifest themselves many generations later.

However, the impression that environmental energies associated with radiation are totally malignant has been emphasized too much; the beneficial uses of radiation have thus been minimized or ignored.

By the proper control of various forms of energies, it is possible to change a device, structure, or material from its original condition into something different. This change can be constructive, rather than destructive, and valuable, instead of detrimental; it can even be life-giving rather than death-dealing; and it can result from nuclear changes as well as chemical changes.

Table VI-3 COMMERCIAL FIRMS LICENSED TO RECEIVE AND DISPOSE OF RADIOACTIVE WASTES

Bay Cities Transportation Co. Pier No. 3 San Francisco 11, Calif.	Nuclear Engineering Co., Inc. Box 146 Morehead, Ky.	Long Island Nuclear Services Corp. Station Road Bellport, N. Y.
California Salvage Co. 700–745 North Pacific Avenue San Pedro, Calif.	Allied Crossroads Nuclear Corp. 201 Victory Road Dorchester, Mass.	Radiological Service Co., Inc. 811 West Merrick Road Valley Stream Long Island, N. Y.
Nuclear Engineering Co., Inc. Box 594 Walnut Creek, Calif.	Laboratory for Electronics, Inc. 1601 Trapelo Road Waltham, Mass.	Nuclear Fuel Services, Inc. Box 124, West Valley Cattaraugas County, N. Y.
U. S. Nuclear Corp. 801 North Lake Street Burbank, Calif.	New England Tank Cleaning Co. 135 First Street Cambridge 41, Mass.	Industrial Waste Disposal Corp. 5420 Calhoun Street Houston, Tex.
The Walter Trucking Co. 1283–1285 East Street New Britain, Conn.	Nuclear Engineering Co., Inc. Box 356 Beatty, Nev.	American Mail Line, Ltd. 740 Stuart Building Seattle 1, Wash.

As long as we apply these energies intelligently, the beneficial effects can far outweigh the deleterious effects. To this end, the safe handling of radioisotopes in production, transportation, utilization, and disposition is constantly directed.

An Analysis of Accidents with Radioisotopes, 1945–1962

By Doris J. Yates*

If accidents with radioisotopes are to be prevented, past accidents should be studied and analyzed for their causes. All accidents involving radiation that occurred in AEC facilities from 1945 through 1962 have been summarized by D. F. Hayes in AEC Report TID-5360 and supplements.[38–42] Typical accidents from these compilations in which radioisotopes were involved are listed in Table VI-4. Reactor and criticality accidents and incidents involving source and special nuclear materials such as natural uranium and thorium, ^{233}U, ^{235}U, and plutonium have been excluded; however, seven accidents with fission products at the production plants at Hanford Works and the Savannah River Plant have been included since they are of the same type that might happen in radioisotope work.

Nearly all accidents listed in the table can be attributed to one or more of the following causes:

1. Failure to adhere to prescribed safety measures
2. Ignorance of the presence of unshielded or excessive radiation
3. Failure of equipment or safety devices

The first and second categories both involve human, as opposed to mechanical, failure; therefore we have combined and analyzed these accidents under the general heading "the human element." This discussion will be followed by a brief description of accidents in which the failure of equipment was the main cause.

The Human Element

The very nature of radioactivity—its invisibility, imperceptibility—makes it easy for the worker to forget that the material he is handling is potentially dangerous; thus most accidents that have occurred with radioisotopes have been the result of human oversight rather than mechanical failure. In nearly all reported instances, the person involved in the accident either neglected safety practices or performed his duties while oblivious to the presence of dangerous material. If we are to learn from the past, we conclude that this neglect or lack of care by the worker is the most likely cause of accidents with radioisotopes.

*Student at University of Tennessee, Knoxville, Tenn., and summer employee at ORNL.

A most striking example of negligence was one of the first accidents recorded by Hayes.[38] In May 1948, four individuals involved in Operation Sandstone, an overseas weapons test, were assigned to collect samples of the cloud resulting from the detonation of an experimental nuclear device. Their duties involved steering drone aircraft equipped with air filters through the cloud and then immediately recovering the contaminated filter papers for radiochemical analysis. Because of wind interference, the men abandoned the 2-ft tongs they had been instructed to use in removing the filter papers and handled the papers with cotton gloves over rubber gloves. One man even abandoned his cotton gloves when they became contaminated. Remarkably, a nearby health-physics man apparently did not object

Table VI-4 SUMMARY OF ACCIDENTS INVOLVING RADIOISOTOPES, 1945–1962

Year	Location	Incident	Injury or loss	Radiation dose	Ref.
		Human Failure			
1948	Pacific weapons-test site	Improper handling of fission-product samples	Radiation burns to hands requiring skin grafts	1.7 to 17 r whole body; no good estimate of dose to hands	38
1957	Houston, Tex.	Release of ^{192}Ir	Widespread contamination; minor radiation burns	1.7 to 3.9 rems during the accident	40
1958	Mound Laboratory	Inhaled ^{210}Po while counting an odd-sized source	Employee received body burden 3.5 times the maximum permissible continuous body burden	Not estimated	40
1949	Los Alamos	Picked up source holder by radioactive end	Overexposure of hand (no visible injury)	40 r to hand	38
1952	Argonne National Laboratory	Radium sulfate capsule broke	Widespread contamination; no injuries	Not significant	38
1956	Fort Belvoir, Va.	Cap removed from a 32-curie ^{192}Ir source container	15 to 25 workers exposed to gamma rays (no detectable injury)	Maximum 6 r	39
1961	Hanford Works	Nine employees contaminated when radioactivity was released	Contamination of local area and workers	Not significant	42
1961	Savannah River	Technician picked up piece of irradiated fuel metal	Exposure of hand to radiation	20 rads to hand	42
1959	Mound Laboratory	Maintenance workers remained in contaminated area too long	Workers received internal body burdens of ^{210}Po about twice the maximum permissible continuous body burden	Not estimated	41
1956	Los Alamos	Worker picked up contaminated tool	Burn on hand required skin grafts	Not given	39
1956	Construction contractor, Milford, Conn.	Worker picked up ^{60}Co source and carried it in glove compartment of car	No detectable effect except two small burns	22 to 26 rads; 3600 r to two small skin areas	39
1959	University of California Radiation Laboratory, Berkeley	Overpressure in a box containing helium caused blowout and release of ^{244}Cm	Widespread contamination; decontamination cost about $30,000	None given	41
		Equipment Failure			
1958	Los Alamos	Failure of a piston ring in a pump resulted in release of ^{3}H to laboratory exhaust system	None	Concentration in room did not go above background	40
1959	ORNL	Repair of fan dislodged particles that were discharged into stack when fan was restarted	Contamination around stack	None	41
1961	Savannah River	Release of ^{131}I from plant stack	No exposure above maximum permissible level	None	42

(Table continues.)

Table VI-4 (Continued)

Year	Location	Incident	Injury or loss	Radiation dose	Ref.
		Equipment Failure (Continued)			
1962	Savannah River	A compression fitting failed, causing a leak of radioactive gas	No detectable area concentration	None	42
1962	Savannah River	A leak developed around the packing of a valve	No detectable area concentration	None	42
1953	Hanford Works	Pieces of irradiated aluminum were washed out of a reactor and handled	Workers received over the permissible radiation dose	30 to 330 rads to hands; 2 to 3.5 r whole body	38
Not given	Not given	Neutron sources leaked	Widespread contamination; in each of two cases, decontamination cost over $30,000	Several people received greater than maximum dose	38
1961	Hanford Works	Condensate lines in chemical processing plant burst	Some area contamination; decontamination cost $2000 to $3000	None	42
1961	Mound Laboratory	Worker dropped and broke contaminated glassware (the glassware was supposed to have been decontaminated)	Contamination of laboratory building	Worker received 1.4 times the maximum permissible continuous body burden of ^{210}Po	42
1957	University of California Radiation Laboratory, Berkeley	Worker held centrifuge tube containing 85 mc of ^{90}Sr	Hand was blistered	1000 to 2000 rems to hand	40
1955	Knolls Atomic Power Laboratory	Radioactive sodium burned in a hot cell	None	Slight	38

to such a deviation from the established procedure. All four men received beta-ray skin burns that required numerous skin grafts. Simply following instructions could have saved four pairs of hands from permanent damage.

A laboratory in Texas was closed down in March 1957 until it could be decontaminated after a small quantity of ^{192}Ir was released into the plant ventilating system.[40] The accident occurred when a sealed can containing 10 aluminum-^{192}Ir pellets was opened. Two $\frac{1}{8}$- by $\frac{1}{8}$-in. pellets, each containing 35 curies of ^{192}Ir, crumbled onto a paper mat on the floor of an open-top hot cell, allowing some particles to become airborne. Radioactive material was carried on employees' street clothes and shoes into their homes and automobiles primarily because (1) the employees ignored a radiation monitor in the laboratory that indicated airborne activity, (2) one person in the laboratory at the time of the spill was wearing street clothes instead of protective clothing, and (3) the contamination was not noticed by officials for almost 1 month after the accident. The widespread contamination required extensive decontamination of both the laboratory and the employees' homes and automobiles. This accident, which was probably the most publicized radioisotope accident to date, would not have attained the magnitude that it did if the men involved had followed the established rules and procedures.

Conscientiousness at the expense of safety can be dangerous! One man received an estimated dosage of 3.5 times the maximum continuous body burden when he accidentally inhaled ^{210}Po from a sample he was counting.[40] The author of the report comments, "The source was odd-sized, and it was necessary to count it in an unorthodox manner." The use of the word "unorthodox" suggests that some of the recommended safety precautions were violated.

Human error can never be completely eliminated, but visible warnings can be used as memory refreshers as they were at Los Alamos Scientific Laboratory in 1949 (Ref. 38). An employee accidentally grasped the "hot" end of a 5-ft aluminum holder containing a 200-mc radium source, which was leaning against the side of a building. He forgot that he had placed it upside down because of snow on the ground. Gamma dosage to the hand was estimated at 40 r, but he suffered no detectable injury. Action was taken to avoid repetition of this error: the hot end of the holder was painted a distinctive color, and the cold end was provided with a handle similar to a file handle. As fur-

ther insurance against repetition, a description of the incident was widely circulated among all employees using similar tools.

An incident at Argonne National Laboratory in 1952 (Ref. 38) points out that attempted helpfulness may be costly. When a 50-mg radium sulfate source used to calibrate pocket meters was broken, unauthorized personnel entered the building to assist in decontamination and subsequently tracked the material through three wings of the building. This unauthorized initiative increased decontamination expenses and could have been extremely serious if more activity had been released.

The fact that radiation cannot be detected by the physical senses is a hazard. The strictest safety measures cannot prevent a few workers' misjudgment of the "hotness" of a substance. Often one may be totally unaware that he has been exposed unless he is carrying a pocket dosimeter. For example, the cap was accidentally pulled out of a "pig" containing a 32-curie iridium radiography source being used for inspecting welds at Fort Belvoir, Va., in 1956. Fifteen to 25 workers were exposed to doses up to 6 r each of gamma radiation before the error was noticed.[39] In this incident, one of the workers had not stored the source properly.

At Hanford, in 1961, construction workers were exposed while removing a plastic "greenhouse" that enclosed a hood over a contaminated conveyor.[42] In removing the tape used to seal the greenhouse to the side of the conveyor, they inadvertently removed the old tape underneath it which covered holes in the conveyor, thus exposing themselves to the contamination. The work area and greenhouse had been surveyed, and no contamination had been detected inside or outside before the crew entered and removed the tape. In the same year, a similar event happened at the Savannah River Plant[42] as part of a periodic hot-cell decontamination and reassembly program. A technician picked up an irradiated black metal fragment, which he thought was a rubber-finger pad for the manipulator, and held it for about 3 sec. He promptly dropped it when he suspected its true identity, but his hand received a dose of about 20 rads.

Still another example of misjudgment occurred at Mound Laboratory in 1959 when two sheet-metal workers spent too much time on a job in a radiation zone.[41] While performing the assigned task of changing supply and exhaust lines on a group of highly contaminated closed hoods, they received internal doses of ^{210}Po approximately twice the permissible maximum continuous body burden in their laboratory.

Los Alamos was the site of an incident similar to others mentioned above involving hot-cell decontamination in 1956 (Ref. 39). A tool used for preparing neutron sources was removed from the cell on a long pole, placed inside a bucket by hand, and taken to the sink for decontamination. Noticing that his pocket dosimeter was off scale, the employee immediately confirmed the dosimeter readings at a monitoring station. Beta burns to the hand were serious enough to require skin grafts and 14 days of hospitalization.

Some workers just cannot resist picking things up. In 1956, a construction worker in Milford, Conn., removed an encapsulated 1600-mc ^{60}Co source, being used to radiograph welds, from a string by which it was suspended. He slipped it into his shirt pocket and then transferred it to the glove compartment of his automobile.[39] He received an estimated whole-body gamma dose of between 22 and 26 rads and approximately 3600 r to each of two small skin areas. Fortunately, no latent effects or blood changes have been detected.

Our final illustration of human failure was caused by pure forgetfulness on the part of a worker. This accident, which took place in Berkeley, Calif., in 1959 (Ref. 41), proved extremely expensive, although equipment damage was slight—a situation that is relatively common in radioisotope accidents. Two experimental target foils each containing 150 μg of ^{244}Cm (10^{11} dis/min) were to be bombarded in a helium atmosphere by a degraded heavy-ion beam. As the air in the target chamber was being replaced by the helium atmosphere, the experimenter forgot to open a valve necessary for the recirculation of the helium. The box became overpressurized, and one of the windows through which the beam was to pass (nickel foil 0.0001 in. thick across a $\frac{3}{8}$-in.-diameter hole) blew out. The curium target immediately exploded, filling the accelerator room with alpha-emitting curium dust. Various costs related to this spill totaled over $30,000.

Failure of Equipment and Safety Devices

When handling or using radioisotopes, one must take into account the possibility of the

failure of laboratory apparatus and safety devices. The most frequent type of isotope incident to date has been the release of radioactive gases or particles into a stack.[39–42] This release usually results from valve leaks or faulty ventilation. Although no release has yet caused serious damage to personnel or surrounding areas, the frequency of this type of accident has dictated that more careful controls be placed on stacks.

Leaks in pipelines and source shielding rank second among the common contamination accidents in radioisotope work.[38,40,42] Breakage of contaminated glass has caused at least one serious laboratory contamination incident,[42] and penetration of wires or other sharp materials through safety gloves worn by electricians and glove-box technicians has also resulted in a few minor burns.[40]

More serious was the spontaneous combustion of radioactive sodium powder which broke out in a hot cell at Knolls Atomic Power Laboratory in Schenectady, N. Y., in 1955 when a leak developed in the apparatus.[38] Fortunately, the possibility of the occurrence had been foreseen and was controlled with minimal effort.

Yet these accidents involving equipment and devices contribute only a minor part to the accident picture when compared to the human factor.

Conclusions

The employer can do a great deal to ensure that his equipment and machinery are in good operating condition, but maintenance of his employees' minds and safety attitude is considerably more difficult.

The following practical rules for safety orientation of employees are suggested by a review of the accidents presented above:*

1. Workers with radioisotopes should respect radiation as they would an electric fence, by keeping their distance whenever possible.
2. Along with the control of the distance factors, exposure times should be carefully controlled.
3. Employees unaccustomed to working with or near radiation should be very thoroughly briefed about where it will be located, how it is to be used, and what to do in case of an emergency.
4. Protective clothing should always be worn in areas where it has been specified, even by persons who intend to stay in the area only a few minutes.
5. Any known contamination or unusual reading on a laboratory air monitor or other counters should be immediately reported to health physics or other safety personnel.
6. Decontamination should be performed only under the direction of qualified and trained personnel.
7. Any unfamiliar object in a radiation zone should be approached with great caution until its activity can be determined.
8. All employees entering containment areas should have proper dosimeters to allow immediate assessment of any foreseeable situation.
9. All tools normally used in handling "hot" material should be marked as such to help safeguard against their being mistaken for "cold" tools.
10. Any deviation from established laboratory procedure involving radiation should be approached only with utmost caution.
11. Areas restricted to authorized personnel should be clearly marked so that even a visitor could recognize those areas he should not enter. It should be stressed that this keep-out warning *includes emergency situations.*
12. Safety measures are established after a great deal of thought and with a definite purpose in mind; therefore they should never be compromised in the interest of expediency.

*This list of rules is not complete since it includes only situations suggested by the accident tabulation. For complete sets of safety rules, see laboratory manuals on radioisotope work.[43–45]

Selling Radioisotope Safety

As was shown in the above articles, radioisotopes are safer to work with than many other items in everyday use, and the number of accidents involving radioisotopes has been low; however, the fear of radiation still remains high in the public mind. In planning tests with radioisotopes, particularly where material may be released to either the air or streams, the public should be informed and convinced of the safety of the proposed tests. Hull, Fries, and coworkers at the California Research Corporation in Richmond, Calif., have used radioiso-

topes for many years in plant tests with a very high degree of safety.[46] The following account, which tells how they handled the public relations aspects of an experiment using tracers in a river, is reported here since it is typical of their handling of each test. This positive approach could be used by others who are planning similar work.

Following the development of the total-count method[47] for flow measurement and tests of the method in a refinery where the isotopes could be completely contained,[48] Hull and Macomber wished to show that the method could be used to measure the flow of streams and rivers. One of their proposed tests was a study of the flow of the American River in the area between Sacramento, Calif., and Lake Tahoe. The U. S. Geological Survey has a number of gaging stations on this river so that it was an ideal place to make intercomparisons between traditional methods and isotopic methods for flow determination. The American River is a rapidly flowing stream originating in the mountains just west of Lake Tahoe and flowing southwest, joining the Sacramento River just north of Sacramento. Since the water supply for Sacramento is taken from the Sacramento River below this juncture, any isotopes put into the American River could conceivably enter the drinking water of the city of Sacramento. Folsom Lake, on the river between the point where the proposed tests were to be made and the confluence of the American and Sacramento rivers, served as a holding and mixing basin.

The first step in selling the safety of the tests was to calculate the levels of radioactivity that would be required. Gold-198 ($t_{1/2}$ = 64 hr) was to be used as the tracer in the tests,[49] and about five injections of 1 curie each would be needed for the series. When these levels had been established and the experimenters were satisfied that there were ample safety margins, an interpretation of the safety factors was prepared, and comparisons were made with common everyday exposures to radioactivity such as those obtained from a radium-dial wrist watch or cosmic rays. (For higher levels of radioactivity, comparisons with exposures in chest or dental X rays could be used effectively.) These calculations are summarized in Table VI-5. In discussions of the concentration of tracers in liquids, the amount of the liquid that a person would have to drink to get the maximum permissible single intake (MPSI) as defined by the AEC was a particularly effective number. In this case, it turned out that one would have to drink 60 gal of water to receive the MPSI.

Table VI-5 SAFETY FACTORS IN ^{198}Au TESTS IN AMERICAN RIVER*

I. At Point of Measurement

A. Drinking water

Maximum concentration will be 8×10^{-5} μc/ml. AEC tolerance for continuous exposure (MPC) is 3×10^{-4} μc/ml.
Maximum permissible single intake (MPSI) for ^{198}Au is 18 μc. (8×10^{-5} μc/ml × 3785 ml/gal = 0.3 μc/gal; MPSI in 18/0.3 = 60 gal.)

B. Swimming exposure

10,000 counts per test is equal to 0.0025 mr.
A wrist watch with 1 μc of radium on its dial gives 1 mr/hr, or 0.0025 mr in about 10 sec.
Cosmic rays give 0.01 mr/hr, or 0.0025 mr in 15 min.

II. At Folsom Dam

The gold is expected to settle in dispersed metallic form on the bed of Folsom Lake. Any that does not settle will be filtered out in the water-treatment plant. But, if both of these processes were to fail, the worst that could happen is calculated below.

A. Maximum concentration

1. Reduced 10-fold by longitudinal mixing
2. Reduced by decay

Assuming 10 days, by a factor of 13,
8×10^{-5} μc/ml ÷ 10 ÷ 13 = 6×10^{-7} μc/ml × 3785 = 2.2×10^{-3} μc/gal.
MPSI in $18/2.2 \times 10^{-3}$ = 8000 gal.

B. Total tolerance for 1 year, AEC basis for averaging, 3×10^{-4} μc/ml × 28,300 ml/gal × 10,000 gal/sec × 3×10^{7} sec/year = 3×10^{12} μc or 3×10^{6} curies. We propose to add only 5 curies in these tests, and this will decay to 0.4 curie in 10 days.

III. Summary of Factors

Maximum hazards	Proposed	Allowable	Safety factor
A. At point of test			
1. In 1 gal of water, μc	0.3	18	60
2. Radiation exposure, mr	0.0025	60	24,000
B. At Folsom Dam			
1. In 1 gal of water, μc	2.2×10^{-3}	18	8000
2. Number of curies averaged over 1 year	0.4	3×10^{6}	7×10^{6}

*The various AEC tolerances that are given were those in effect in 1958. Some of these have changed slightly in the past 6 years.

The next step was to sell the proposal in person to the various regulatory agencies involved. The proposal was first taken to the California Department of Public Health which, in turn, cleared it with the Division of Industrial Safety, the Fish and Game Commis-

sion, the Department of Water Resources, and the District Engineer. Next, the information was taken to the Sacramento County Health Officer and the Central Valley Water Pollution Board. The proposal was explained in detail, and all questions were answered; the researchers always emphasized their desire to give full information. The officials tentatively approved the proposal, but made it contingent upon a favorable public response to a notice announcing the tests. (The officials were particularly worried at this time because there had just been a great deal of public discussion about fallout radioactivity in the lakes and rivers in northern California.)

The next step was the preparation of the news release, which emphasized the benefits expected from the tests and also stressed the word "safety" rather than "hazard"; however, no attempt was made to play down the radiation problem, only to present it in proper perspective. A happy coincidence that was used in preparing this release was the fact that the American River flows near Sutter's Mill, where gold was first discovered in California. The news release follows:

PLACERVILLE, June 24, 1957—In a switch on the old '49er gold-panning technique, California Research Corporation scientists will "salt" the historic American River near here with "atomic" gold to measure the volume of water flow in the fast-moving stream.

The test, scheduled for tomorrow and Wednesday, will in no way affect the safety of the river water for drinking, swimming or other use, company and county health officials emphasized. California Department of Public Health representatives will also supervise the test to assure that safety standards are maintained.

The new radioactive testing method, designed for use in petroleum refineries and adapted to stream measurement, is expected to find important applications in the planning of flood control and irrigation programs and in mapping watershed run-off.

Developed by the Standard Oil Company of California research subsidiary, the new method involves adding a small amount of radioactive gold isotope to the river, then recording the flow of the gold particles with a Geiger counter about 1½ miles downstream.

Quick calculations based on the number of Geiger counter clicks as the gold flows past tell how many cubic feet per second are carried by the stream. Personnel equipped with fishing pole, line, and Geiger counter instead of the traditional hook can complete the test in only an hour or two.

A. L. Lyman, Cal Research president, explained that use of a gold isotope in the test eliminates any radiation hazard—gold quickly loses its radioactivity, compared to some metals which retain it for years.

Dr. Ira Church, county health official, said further that none of the minute particles of gold would reach Sacramento homes. The gold is diluted rapidly and most of it will settle out on the bottom of Folsom Lake. Filtration in the water treatment plant will remove any traces left after that.

Known as the "total-count" method, the new Cal Research technique promises to save time and labor in surveying water resources in regions not equipped with gauging stations, which require dams or weirs to gather water-flow data.

Cal Research plans to make the technique and necessary instructions available for general use under license.

After the story was given to the paper, the research team and local health officers waited for objections. A day after the newspapers came out, not a single call or question had been received. Therefore the county health officer approved the tests, and they were run. During each test no restrictions were put on swimmers except to prohibit swimming between the point of tracer injection and the point of counting. Because the temperature was about 110°F where they were working, Fries swam in the water at the counting point (Fig. VI-8). Fries said he had no qualms about the radioactivity because his exposure while the whole 1-curie tracer wave went by would not exceed 0.0025 mr. This photograph was released to the newspapers to demonstrate the safety of

Fig. VI-8 B. A. Fries swims in the American River during a flow measurement with ^{198}Au, while D. E. Hull counts the radioactivity.

the test. The drinking water in Sacramento was monitored for 2 weeks following the tests, but no activity was found.

The newsworthy aspect of the test prompted the Associated Press to pick up the story, and they distributed it far and wide; clippings telling about the tests were received from as far away as Panama. Many used the story as background for a feature article. The first test was so successful that the next year, when further tests were planned,[50] approval was obtained from the Department of Public Health and other agencies involved merely by sending them a letter telling them that the test was scheduled and inviting any interested persons to attend.

This case history shows that a positive approach in the public relations aspects of radioisotope work can be very valuable. A timorous public relations manager might have seen only the possibility that the company would be criticized for poisoning the water with "that terrible radioactive stuff." The California Research public relations man, on the other hand, took the positive view — radioactivity can make a good story when it is told right. The more often radioisotope work is presented in this light, the faster these tools will be accepted by the public.

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Section

VII

Isotopes and Radiation Technology

Miscellaneous Items of Interest

Conferences

Symposium on Low-Energy X and Gamma Sources and Applications

An interesting 2-day meeting on the above subject was held at the Illinois Institute of Technology Research Institute (IITRI) on Oct. 20 and 21, 1964. The program was listed on page 96 in Vol. 2(1) of this review. The papers will be published as proceedings of the meeting; however, a few items are mentioned at this time.

After welcoming remarks by Leonard Reiffel of IITRI and an outline of programmatic objectives by J. W. Hitch of the AEC Division of Isotopes Development (DID), a number of investigators reported on their researches dealing with secondary X-ray and bremsstrahlung sources.

J. J. Ezop and *T. G. Stinchcomb* of IITRI listed some of the advantages of such sources, including portability, compactness, stability, freedom from auxiliary power and maintenance, and relatively low cost. Although the conversion efficiency is generally less than 1%, they estimate that it could be as high as 20% with very heavy targets. IITRI has been working with ^{147}Pm, ^{85}Kr, and ^{90}Sr, using tin and lead matrixes with various geometric arrangements (90 and 180° reflection, sandwiches, etc.).

R. S. Pressly and *F. N. Case* of the Oak Ridge National Laboratory (ORNL) described the results of their work with ^{125}I (a true X-ray source), ^{147}Pm, ^{90}Sr, ^{151}Sm, ^{99}Tc, ^{55}Fe, and ^{3}H, showing radiographs taken with several of the sources. *Charles Badgett* of Industrial Nucleonics reviewed a number of applications of secondary sources in industry, and *Thomas Bustard* and *V. C. Truscello* of Martin reported separately on the bremsstrahlung from ^{90}Sr and ^{147}Pm in thin absorbers.

J. F. Cameron of the International Atomic Energy Agency (IAEA) surveyed the applications of low-energy X and gamma radiations in Europe and suggested that the one advantage of the small size of such sources was the possibility of in-line use. He gave a number of specific uses involving X-ray fluorescence, transmission, and backscatter and pointed out that the matrix-absorption technique could be quite helpful in many instances.

In the medical session on October 21, *R. S. A. Söremark* of Harvard University showed how ^{125}I can be used satisfactorily for intraoral X radiography. The definition resulting from the use of point sources is good, and accessibility to difficult locations is simplified. Iodine-125, electrodeposited on copper wire with 85% yield and covered with epoxy, gives a very satisfactory source. The absorbed dose from a 200-mc ^{125}I source for 60 sec is much less than that required from a conventional X-ray machine to take the same picture. This is the case partly because the maximum sensitivity of the film is at about the energy of the ^{125}I radiation, i.e., 0.03 Mev.

Lawrence Lanzl and *Nels Strandjord* of the University of Chicago described a technique for the nondestructive studies of bone mineral content using ^{125}I. Then *John Cameron* and *James Sorenson* of the University of Wisconsin described the application of low-energy isotopes such as ^{125}I for determining the ratio of muscle to fat in studying body composition. Their work is based on the observation that the ratio of absorption, muscle to fat, changes rapidly from 0 to 0.06 Mev. Thus, by use of two different energies of radiations, it is possible to get two different ratios of muscle to fat.

There were other very excellent papers presented, and, in general, the consensus seemed to be that there is a bright future for this area of application and that an almost limitless range of energies (characteristic X ray, bremsstrahlung, etc.) can be made available by suitable choice of radioisotope or radioisotope and matrix.

Clinical Uses of Nuclear Medicine Subject of Symposium in Ohio

A symposium on clinical application of nuclear medicine was held Oct. 30 and 31, 1964, at Shaker Heights, Ohio. Picker X-Ray Corporation and E. R. Squibb & Sons sponsored the meeting; the program was as follows:

Radiopharmaceuticals

New Radiopharmaceuticals	Paul Numerof, Squibb Institute of Medical Research, New York
The Short-Lived Nuclides	William G. Myers, Ohio State University, Columbus
Applications of Technetium-99*m*	Katherine A. Lathrop, Argonne Cancer Research Hospital, Chicago
Micro and Macroaggregates of Albumin	George V. Taplin, University of California Medical Center, Los Angeles
The Future of Nuclear Medicine and Radiopharmaceuticals	Merrill A. Bender and Monte Blau, Roswell Park Memorial Institute, Buffalo

Thyroid Studies

Thyroid Disease	A. Stone Freedberg, Beth Israel Hospital, Boston
Thyroid Scanning	J. Martin Miller, Henry Ford Hospital, Detroit
The T-3 Test	Milton W. Hamolsky, Rhode Island Hospital, Providence

Cardiovascular Studies

Isotope Applications to Hematology	Major Marcel E. Conrad, Walter Reed Army Institute of Research, Washington
Blood Volume Determinations	Solomon N. Albert, Washington Hospital Center, Washington
Iron Turnover Investigations	Major Marcel E. Conrad
Cardiac Output Measurement	Kenneth R. Kaess, St. Mary's Hospital, Waterbury, Conn.
Cardiac Scanning	N. David Charkes, Albert Einstein Medical Center, Northern Division, Philadelphia

Renal Studies

Renal Blood Flow	Major Gerald S. Johnston, Walter Reed General Hospital, Washington
The Renogram	George V. Taplin
The Renal Scan	N. David Charkes

Liver, Spleen, Brain, and Tumor Studies

Liver Scanning	James Christie, Euclid-Glenville Hospital, Cleveland
Albumin Aggregates for Liver Scanning	George V. Taplin
Liver Blood Flow	Nilo E. Herrera, Danbury Hospital, Danbury, Conn.
Spleen Scanning	Philip M. Johnson, Presbyterian Hospital, New York
Brain Scanning	Bertram Selverstone, New England Center Hospital, Boston
Tumor Scanning	D. Bruce Sodee, Doctors Hospital, Cleveland

Newer Scanning Techniques

Lung Scanning	Gerhard F. Fueger, The Johns Hopkins Medical Institutions, Baltimore
Bone Scanning	N. David Charkes
Pancreas Scanning	D. Bruce Sodee
Parathyroid Scanning	E. James Potchen, Peter Bent Brigham Hospital, Boston

International Dating Conference

The Sixth International Carbon-14 and Tritium Dating Conference will be held June 8–11, 1965, at Washington State University, Pullman, Wash.

The Conference is cosponsored by the Institute of Technology at Washington State University and the AEC Division of Isotopes Development.

About 30 papers will be given, covering:

Improvement in Methods of Carbon and Tritium Dating
New Archaeological Conclusions
Geophysical Implications of Radiocarbon and Tritium Dating
Description of Carbon-14 and Tritium in the Atmosphere and Hydrosphere
Water Dating

Scientists are expected to attend from 25 foreign countries, including New Zealand, India, South Africa, Switzerland, Japan, Norway, New South Wales, Denmark, France, the Netherlands, Sweden, West Germany, Ireland, England, and Canada. The group represents the heads of the major carbon dating and tritium laboratories throughout the world, according to the organizer of the meeting, Roy M. Chatters, Associate Nuclear Engineer at the University and head of the Radioisotopes and Radiations Laboratory.

Announced chairmen of sections are James B. Griffin, Director of the Museum of Anthropology at the University of Michigan, Anthropology and Archaeology Section, and A. G. Schrodt, Scientific Director, Packard Instrument Company, Techniques Section.

The fifth international conference on this subject was held in 1962 at Cambridge, England. [See *Nature*, 4845: 943-5 (Sept. 8, 1962).]

AEC Activities

Kansas Joins List of States Assuming Regulatory Authority

Kansas will assume part of AEC's regulatory authority over the uses of radioactive materials in the state on Jan. 1, 1965.

Kansas is the ninth state to assume this responsibility. Others are Arkansas, California, Florida, Kentucky, Mississippi, North Carolina, New York, and Texas.

More Labeling Data Required Under New Proposed Amendment

The AEC is considering an amendment to require more information on containers holding licensed radioactive material.

Under the new rule, containers of above-specified quantities would be labeled to show the kinds of radioactive material inside, the approximate activity, and the date on which the activity was determined. Presently these data are required only on labels of storage containers. The proposed amendment would extend the requirement to containers in which the material is used or transported. Natural uranium or thorium are excluded.

Notice of the proposed amendment to 10 CFR Part 20 was published in the *Federal Register* on Oct. 29, 1964. Sixty days are allowed for public comment.

SNAP Mockup Shot to Bermuda from Virginia in Dark of Moon

A mockup of a SNAP isotopic power generator was launched in the dark of the moon on Oct. 9, 1964, from Wallops Island, Va. The object of the test, designated Reentry Flight Demonstration No. 2 (RFD-2), is to acquire data on isotopic-power-generator disassembly and fuel-capsule burnup rate for use in advanced generator designs. The 800-mile suborbital flight was at 12:05 a.m. to aid visual observation of brightly burning tracer elements in the nonradioactive fuel capsules. The mockup was launched by a Scout rocket from the National Aeronautics and Space Administration's facility to an impact point southeast of Bermuda.

Test Explosion for Isotopes

In a program to develop a nuclear device for producing isotopes, the AEC conducted a 20-

to 200-kt underground test on Oct. 9, 1964, at the Nevada Test Site. The method sought would be capable of producing neutron-rich isotopes of known transplutonium elements and possibly new, heavier elements.

California Nuclear Licensed To Use Land Near Richland

The AEC recently issued the first license to a private nuclear firm for use of land leased by the Commission to the State of Washington near Richland, Wash. California Nuclear, Inc., Lafayette, Ind., received the license for receipt, processing, repackaging, and storage of radioactive waste material that it receives from its customers. The company will sublease 100 acres of land from the state.

The company plans to construct two buildings. One building will have offices, a health-physics counting room and laboratory, maintenance facilities, and room for storage of nonradioactive materials. A second building will be used for solidification of liquids, decontamination, and storage. Radioactive waste will be stored within a fenced area.

California Nuclear will be authorized to possess up to 50,000 curies of radioisotopes, 4000 lb of uranium and thorium, and 5000 g of special nuclear material.

AEC, Contractor Negotiate Boron-10 Plant Agreement

The AEC and Nuclear Materials & Equipment Corporation, Apollo, Pa., are negotiating a contract to operate an AEC ^{10}B production plant near Niagara Falls, N. Y. The plant has been on standby since 1958. AEC recently found that current inventories could not meet future demands, primarily those for reactor development.

Thirteen firms were invited to submit proposals, and five bids were received. About 70 people will operate the plant, with an annual operating cost of about $1 million. A cost-plus-fixed-fee contract would extend through Dec. 31, 1967.

Space Generator Tested at ORNL

The Radio Corporation of America has finished testing a space generator at ORNL in an AEC program to develop technology involving radioisotopic power for space vehicles. The generator, similar to the SNAP systems, will be fueled with approximately 80,000 curies of ^{90}Sr.

ORINS Announces Schedule in Advanced 1965 Program

ORINS has announced the 1965 schedule for an expanded program of courses designed to train personnel in the safe and efficient use of radioactive materials and to stimulate further interest in their applications.

The courses are presented by the ORINS Special Training Division for the AEC.

The calendar of courses for 1965 includes:

Four-week basic courses on the use of radioisotopes in research, January 11–February 5, March 22–April 16, May 3–28, August 9–September 3, and September 27–October 22.

Three-week medical-qualification courses for physicians which fulfill the minimal requirements for licensure by AEC to use radioisotopes in diagnostic procedures, January 4–22, March 1–19, May 3–21, and September 13–October 1.

Ten-week health-physics course on the study, evaluation, and control of radiation hazards, September 13–November 19.

Three-week advanced health-physics courses for more specialized training in recent developments and new techniques in the field, March 29–April 16, and November 29–December 17.

Mobile Isotope Laboratories To Visit 25 College Campuses

Two mobile radioisotope laboratories will visit 25 college campuses in 1965.

The Oak Ridge Institute of Nuclear Studies (ORINS) operates these units for the AEC. In the 5 years of its operation, the program has been presented at 73 colleges in 29 states, with approximately 900 faculty members and students participating.

Laboratory experiments and lectures are presented for 2 weeks at each campus. They cover basic counting techniques and applications of radioisotopes in the fields of physics, chemistry, and biology. Six to eight students take part in each 2-hr laboratory session. The schedule follows:

October 5–16, Farmington State Teachers College, Farmington, Maine; October 12–23, Knoxville College, Knoxville, Tenn.; October

19–30, Keene State College, Keene, N. H.; October 26–November 6, Georgia State College, Atlanta, Ga.; November 9–20, Danbury State College, Danbury, Conn., and Jacksonville State College, Jacksonville, Ala.; November 30–December 11, Fairfield University, Fairfield, Conn., and Ouachita Baptist College, Arkadelphia, Ark.; January 4–15, Marymount College, Tarrytown, N. Y., and Mississippi Southern College (University of Southern Mississippi), Hattiesburg, Miss.; January 18–29, Southern University, Baton Rouge, La., and Wagner College, Staten Island, N. Y.; February 1–12, Wilson College, Chambersburg, Pa.; February 8–19, St. Edward's University, Austin, Tex.; February 22–March 5, Mary Hardin–Baylor College, Belton, Tex., and Millersville State College, Millersville, Pa.; March 8–19, Hood College, Frederick, Md.; March 15–26, Bishop College, Dallas, Tex.; March 22–April 2, Howard University, Washington, D. C.; March 29–April 9, Oklahoma College for Women, Chickasha, Okla.; April 5–16, Virginia State College, Petersburg, Va.; April 26–May 7, Mary Baldwin College, Staunton, Va., and Wayland Baptist College, Plainview, Tex.; May 10–21, Sul Ross State College, Alpine, Tex., and West Virginia Wesleyan College, Buckhannon, W. Va.

Advanced medical-qualification course for physicians who have previous experience in the use of radioisotopes and desire more specialized training, April 5–9 (tentative).

The schedule for summer institutes and other special courses that are also regularly offered by the Division will be announced later. Additional information concerning courses, schedules, fees, and application procedures may be obtained from the Special Training Division, Oak Ridge Institute of Nuclear Studies, P. O. Box 117, Oak Ridge, Tenn. 37831.

Availability of Isotopes and Services

Highly Enriched ^{46}Ca, ^{180}Ta Now Available from ORNL

Several hundred milligrams of ^{46}Ca, with purities ranging from 32 to 46% and prices estimated at ~$500 to $700/mg, have recently been placed in inventory at ORNL. Also available is 300 mg of 0.34% ^{180}Ta (natural abundance 0.012%) at $35/mg.

General

USSR Promotes Use of Radioisotopes Through Its "Isotopes" Association

The monthly French publication *Atomes* notes in its November 1964 issue that the USSR is promoting the widespread use of radioisotopes in agriculture, medicine, and industry through an organization, ISOTOPES, founded to develop applications of radioisotope techniques. The organization is housed in a seven-story building in the Soviet capitol, and offices have been set up in other cities to demonstrate basic instrumentation and techniques.

Radioisotopes are used extensively in the USSR in gaging, quality control, automation, and static elimination—for example, in the poultry operation at Tomolino, the Institute of Radiological Medicine at Obninsk, the chrome works at Aktubinsk, and the metallurgical combine at Novotoula.

A cyclotron at Obninsk furnishes radioactive isotopes. In addition, this year at Tiflis stable isotopes will be separated. The publication comments that the activity in the USSR indicates it will be a significant exporter toward 1970.

New Developments in Processing

Several new developments in radiation processing are reported in the following items, including new Food and Drug Administration (FDA) rulings, the opening of the Marine Products Development Irradiator (MPDI), and negotiations on a Novawood facility.

FDA ACTIONS

The FDA recently approved ^{137}Cs as a gamma-radiation source for use on wheat, wheat products, and white potatoes. In this action FDA acted on an AEC petition; previously it had cleared ^{60}Co as a gamma source for wheat and potato irradiation.

FDA also cleared packaging materials for use in the radiation processing of prepackaged foods. Hazleton Laboratories, Inc., Falls Church, Va., had filed the petition on behalf of AEC. Materials cleared are: nitrocellulose-coated cellophane, glassine paper, wax-coated paperboard, and five plastic films. The FDA order allows these materials to be irradiated up to 1 Mrad in the gamma-radiation processing of prepackaged foods.

MPDI DEDICATED

The MPDI was dedicated Sept. 28, 1964, at the Bureau of Commercial Fisheries Technological Laboratory, Gloucester, Mass.

The seafood irradiator, built to demonstrate the feasibility of extending the refrigerated storage life of fresh fishery products, will be operated for the AEC by the Bureau of Commercial Fisheries Laboratory, U. S. Department of the Interior. Irradiated seafood, like that which will be processed by the facility, was served to luncheon guests following the dedication. Guests were served radiation-processed clams, broiled haddock, and lobster.

Principal speakers were Representative William H. Bates of Massachusetts, Mary I. Bunting, AEC Commissioner, Brigadier General Woodrow W. Vaughn, U. S. Army Natick Laboratories, and Charles Butler, Bureau of Commercial Fisheries.

Mayor Ralph B. O'Maley of Gloucester and other state and local officials attended the ceremony, along with officials of the Bureau of Commercial Fisheries, representatives of the fishing, seafood, and allied industries, and scientists who were in Boston attending an International Conference on Radiation Preservation of Foods, Sept. 28 to 30, 1964.

Construction of the seafood irradiator was begun in late July 1963. The $600,000 facility will operate on a near-commercial scale, processing marine products at about 1 ton/hr using a 250,000-curie ^{60}Co irradiation source. [See *Isotopes and Radiation Technol.*, 1(4): 310-7, 1964.]

A few pertinent MPDI facts follow:

The MPDI building is a one-story structure divided into a general area and an irradiation cell. The general area of 2500 sq ft includes a lobby, office, laboratory space for health physics and dosimetry, cold-storage room, filleting area, and conveyor loading area. The irradiation cell has a gross area of 1250 sq ft. Its concrete walls are more than 5 ft thick, and the 4-ft-thick ceiling has a removable plug for lowering a 7-ton lead shipping cask containing the radiocobalt into the storage well. Operations within the irradiation cell are viewed and controlled remotely by television and electrical controls.

The radiation source is raised by an elevator and is placed in a horizontal position inside an aluminum air-cooled shroud between the tracks of the conveyor. The source plaque is approximately 1 by 4 ft, in six subunits. When not in use, the source is stored in 15 ft of water in a stainless-steel well within the irradiation chamber.

There are a number of safety interlocks in the irradiation cell to prevent accidental exposure of personnel to radiation from the source. Strategically placed openings in the walls and roof are used to introduce long-handled tools in the event of elevator malfunction. The irradiation cell is kept at a lower pressure than the rest of the building, with air being drawn from the building into the cell and then exhausted through a filter stack.

The pasteurization process can be introduced with little disturbance to commercial fish-distribution procedures. After the fish has been filleted and packed, the packages are sent through the irradiation cell. Fillets of finned fish are handled in rectangular tins holding 30 lb of product. Shellfish, such as clams, will be packed in commercial No. 10 cans.

In the normal operation of the MPDI, the seafood packages will be brought into the building and placed on movable racks in the cold-storage room. The room has a capacity for 1½ day's supply of incoming irradiated seafood and end product, based on a one-shift operation. As an alternative procedure, the filleting can be done inside the building.

NOVAWOOD NEGOTIATIONS

AEC is negotiating a contract with Vitro Engineering Company of New York to conduct engineering-design studies and cost analyses on a conceptual design for a pilot plant for radiation processing of plastic-impregnated wood. Vitro was selected from among 28 companies that filed proposals to AEC.

The facility to be designed would be used to impregnate a wide range of wood species, sizes, and shapes under various conditions with a wide variety of monomers. The facility's production rate would be about 2000 lb/hr.

Novawood results from impregnating wood with a simple liquid monomer and then exposing the treated wood to radiation from a gamma-ray source such as ^{60}Co. The radiation forms chain polymers within the wood, producing a solid wood-plastic composite. It shows promise for use in floors, window frames, sills and doors, tool handles, indoor and outdoor furniture, structural beams, sporting goods, boat decks and fittings, shoe lasts, and dies and jigs.

Translations Avilable

As part of the effort to cover the foreign literature in the isotopes field, selected articles are translated by members of the ORNL Isotopes Development Center. These are available through usual AEC information dissemination channels (see front cover). Some of the recent

translations of interest in the field of production and applications of isotopes are:

1. ORNL-tr-221: B. N. Petrov, S. N. Bernov, and D. Ya. Libenson, Applications of the Mössbauer Effect, *Vestn. Akad. Nauk SSSR*, pp. 40-3 July 1963 (in Russian; translated by Martha Gerrard and J. J. Pinajian).

2. ORNL-tr-387: V. I. Sinitsyn, Use of Radioactive Isotopes in the Metallurgical Industry of Some of the Soviet Economic Group Countries, *At. Energ.*, 16: 279-82 (in Russian; translated by Martha Gerrard).

3. ORNL-tr-309: P. Morzek, Activation Perturbation by Self-absorption and Flux Depression in Activation Analysis, *Kernenergie*, 5: 839-45 (1962) (in German; translated by A. M. Ehrlich and J. J. Pinajian).

4. ORNL-tr-300: L. N. Moskvin, B. K. Preobrazhenskii, and L. N. Rzhanitsyna, The Use of an Ion Exchange Resin as the Aqueous Phase Carrier in Partition Chromatography. The Separation of Zn, Cd, and Hg, *Radiokhimiya*, 5: 299-304 (1963) (in Russian; translated by S. J. Rimshaw and J. F. Allen).

5. ORNL-tr-175: J. J. Bouquiaux and J. H. C. Gillard, Separation of Alkaline Earths by EDTA on Ion Exchange Resins, *Anal. Chim. Acta*, 30: 273-9 (1964) (in French; translated by J. F. Allen).

6. AEC-tr-6187: K. Wagner and H. Poser, A Study of the Formation of ^{59}Fe from ^{59}Co with Fission Neutrons, *Kernenergie*, 6: 177-8 (1963) (in German; translated by J. J. Pinajian).

7. ORNL-tr-315: V. C. Zolotarev and V. M. Gusev, Electromagnetic Separation of Osmium Isotopes, *Pribory i Tekhn. Eksperim.*, 1964, No. 1: 141-2 (in Russian; translated by G. M. Banic).

8. ORNL-tr-248: Hermann Kuster (inventor), Shipping Container for Radioactive Heat-emitting Materials, Especially for Spent Reactor Fuel Elements, German Patent 1,146,209 (Mar. 28, 1963) (in German; translated by S. J. Rimshaw).

9. ORNL-tr-193: W. Eyrich, The Construction and Use of Neutron Generators, *Umschau Wiss. Tech.*, 63: 695-7 (1963) (in German; translated by J. J. Pinajian).

10. ORNL-tr-209: V. I. Levin, I. V. Meshcherova, and V. K. Semrov, The Isolation of Carrier-free ^{54}Mn from Neutron-irradiated Iron by Solvent Extraction, *Radiokhimiya*, 3: 417-21 (1961) (in Russian; translated by S. J. Rimshaw and J. J. Pinajian).

11. ORNL-tr-124: K. Wagner, Some Investigations on the Production of Carrier-free ^{54}Mn and a Mixture of Carrier-free Cobalt Isotopes ^{56}Co, ^{57}Co, and ^{58}Co from Iron, *Kernenergie*, 5: 853-9 (1962) (in German; translated by J. J. Pinajian).

12. ORNL-tr-247: V. I. Levin and V. V. Bochkarev, The Production of Radioisotopes in a Reactor by Means of Threshold, Consecutive, and Secondary Nuclear Reactions, *Tr. Tashkentsk. Konf. po Mirnomu Ispol'z. At. Energii Akad. Nauk UzSSR*, 2: 368-72 (1960) (in Russian; translated by S. J. Rimshaw and J. F. Allen).

13. AEC-tr-6186: S. Niese, P. Morzek, and C. Herold, Yield Determinations of Threshold Reactions Ti(n,p)Sc and Ge(n,α)Zn in the Water Cooled–Water Moderated Reactor, *Kernenergie*, 6: 37-9 (1963) (in German; translated by J. J. Pinajian).

Selected Current Literature

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J. H. Lawrence, B. Manowitz, and B. S. Loeb, *Radioisotopes and Radiation*, McGraw-Hill, New York, 1964 (131 pp.; price $15.30).

Proceedings of the Conference on Methods of Preparing and Storing Marked Molecules, Presses Academiques Européennes, Brussels, Belgium (price 1000 FB or $20).

F. L. Brannigan, *Radiation in Perspective*, Text of a Film Lecture, Superintendent of Documents, U. S. Government Printing Office, Washington, D. C. 20402 (price 10¢).

R. H. Lafferty, Jr., P. S. Baker, and Martha Gerrard, Publications on Isotopes at Oak Ridge National Laboratory 1946–1964; a Bibliography of ~1700 References with Keyword, Isotope, Report Number, and Author Indexes, USAEC Report ORNL-IIC-1, Oak Ridge National Laboratory, July 1964 (312 pp.; available from Clearinghouse for Federal Scientific and Technical Information, National Bureau of Standards, U. S. Department of Commerce, Springfield, Va.; price $4.00).

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L. G. Stang, Coordinator, Manual of Isotope Production Processes in Use at Brookhaven National Laboratory, USAEC Report BNL-864, Brookhaven National Laboratory, August 1964 (69 pp.; available from Clearinghouse for Federal Scientific and Technical Information, National Bureau of Standards, U. S. Department of Commerce, Springfield, Va.; price $1.75).

A. Oltmann and O. A. Kuhl, Fabrication of BNL Standard Cobalt-60 Source, USAEC Report BNL-848, Brookhaven National Laboratory, February 1964 (23 pp.; available from Clearinghouse for Federal Scientific and Technical Information, National Bureau of Standards, U. S. Department of Commerce, Springfield, Va.; price $0.75).

H. T. Russell, Production and Recovery of Tin-113, Antimony-125, and Tellurium-125*m* from Reactor-irradiated Tin, USAEC Report ORNL-TM-782, Oak Ridge National Laboratory, September 1964 (6 pp.; available from Clearinghouse for Federal Scientific and Technical Information, National Bureau of Standards, U. S. Department of Commerce, Springfield, Va.; price $1.00).

R. E. McHenry and J. C. Posey, An Air-pulsed Laboratory Mixer-Settler, USAEC Report ORNL-TM-896, Oak Ridge National Laboratory, September 1964 (20 pp.; available from Clearinghouse for Federal Scientific and Technical Information, National Bureau of Standards, U. S. Department of Commerce, Springfield, Va.; price $1.00).

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